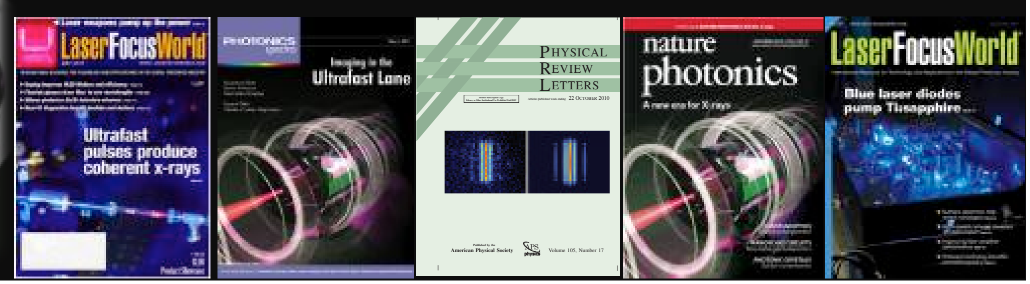
Coherent keV X-Rays from Tabletop Femtosecond Lasers and Applications in Nanometrology



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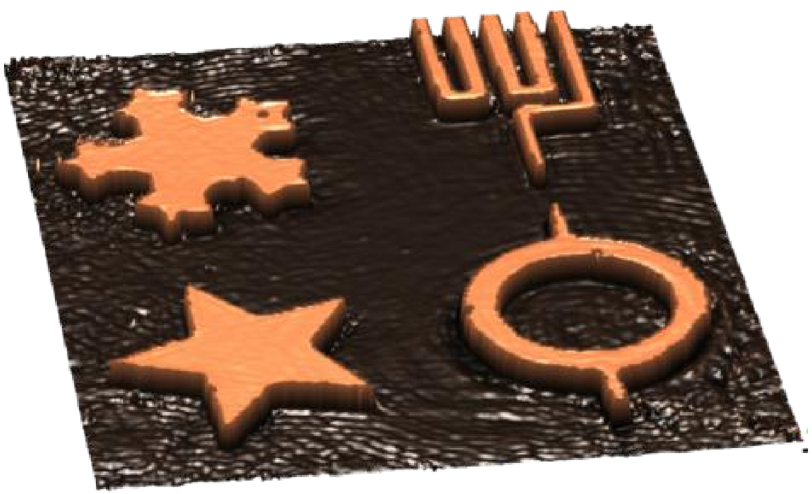
HGST

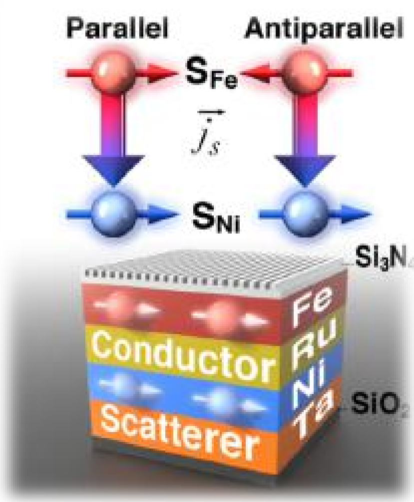
Outline

- **High harmonic generation (HHG)**
  - ONLY tabletop source of coherent soft X-rays
  - Recent advance: bright beams from UV to keV
  - Limits? Hard X-ray beams?
  - Commercial systems in EUV
- **Unique nanometrologies using HHG**
  - Coherent (lensless) imaging near wavelength limit
  - EUV acoustic and thermal nanometrology
  - Photoemission spectroscopy, spintronics, etc

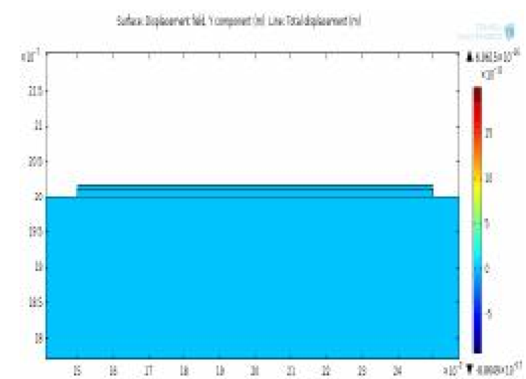
3D Coherent Imaging



Nanoscale spin dynamics

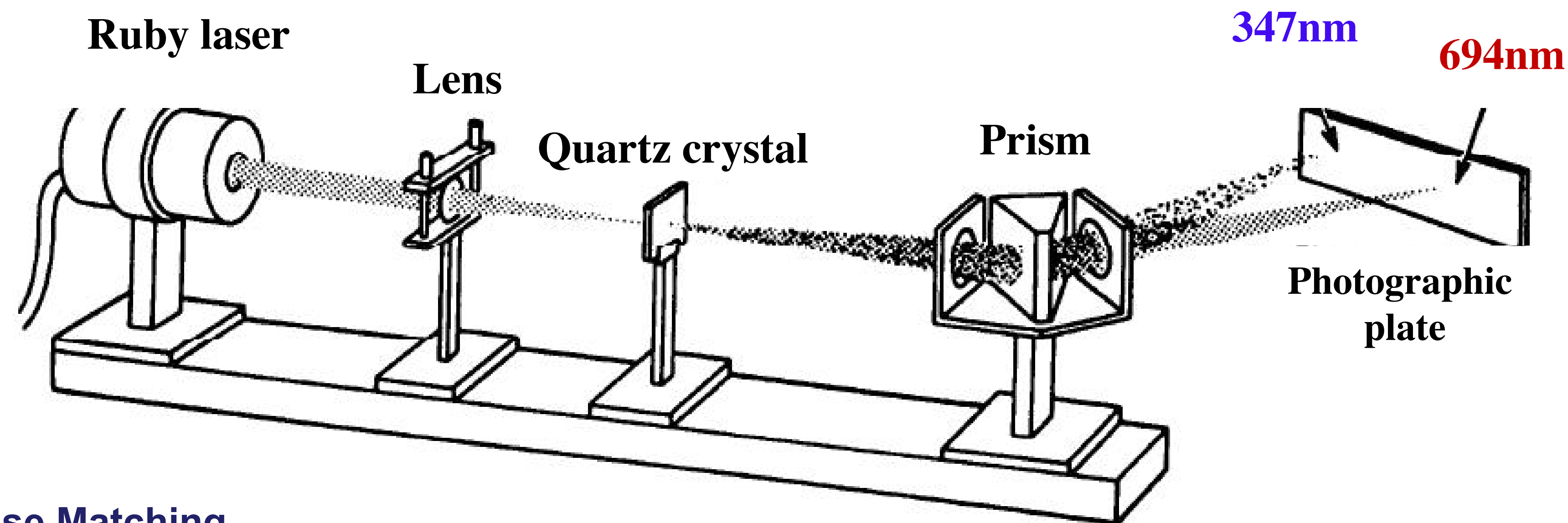


Acoustic/thermal nanometrology

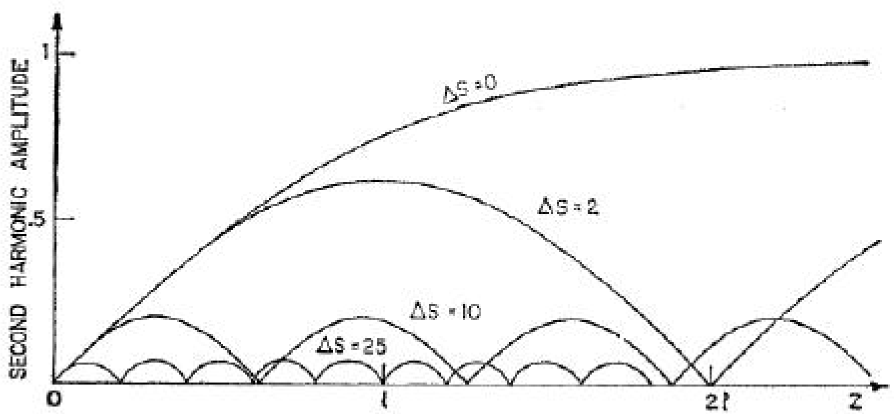


Nonlinear Optics: microscopic and macroscopic science

Second harmonic generation
*Franken et al, PRL **7**, 118 (1961)*



Phase Matching
*Armstrong, Bloembergen et al., PRA **127**, 1918 (1962)*



$$\boxed{V_{phase}(2\omega) = V_{phase}(\omega)}$$

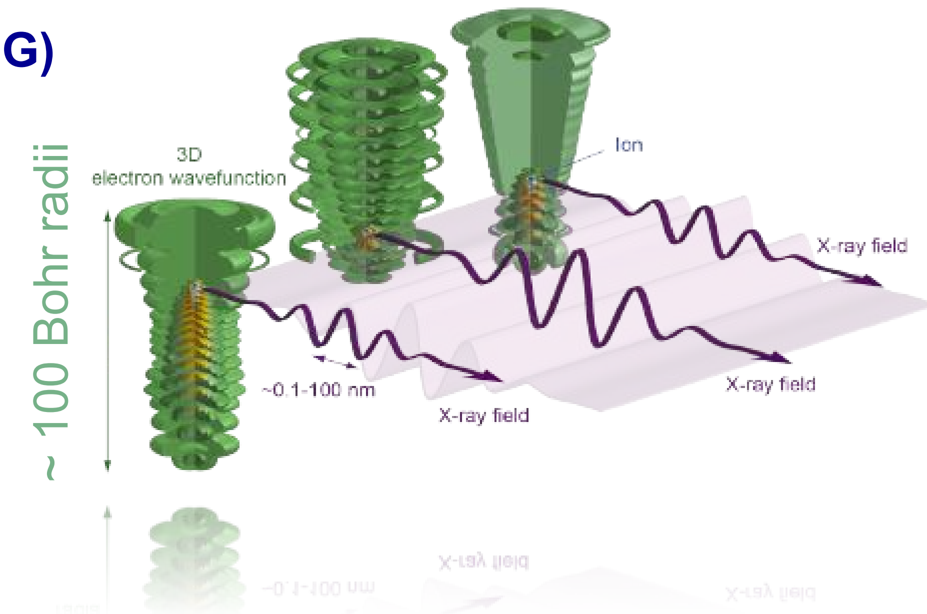
k_ω k_ω

$k_{2\omega}$

Extreme nonlinear optics: microscopic and macroscopic science

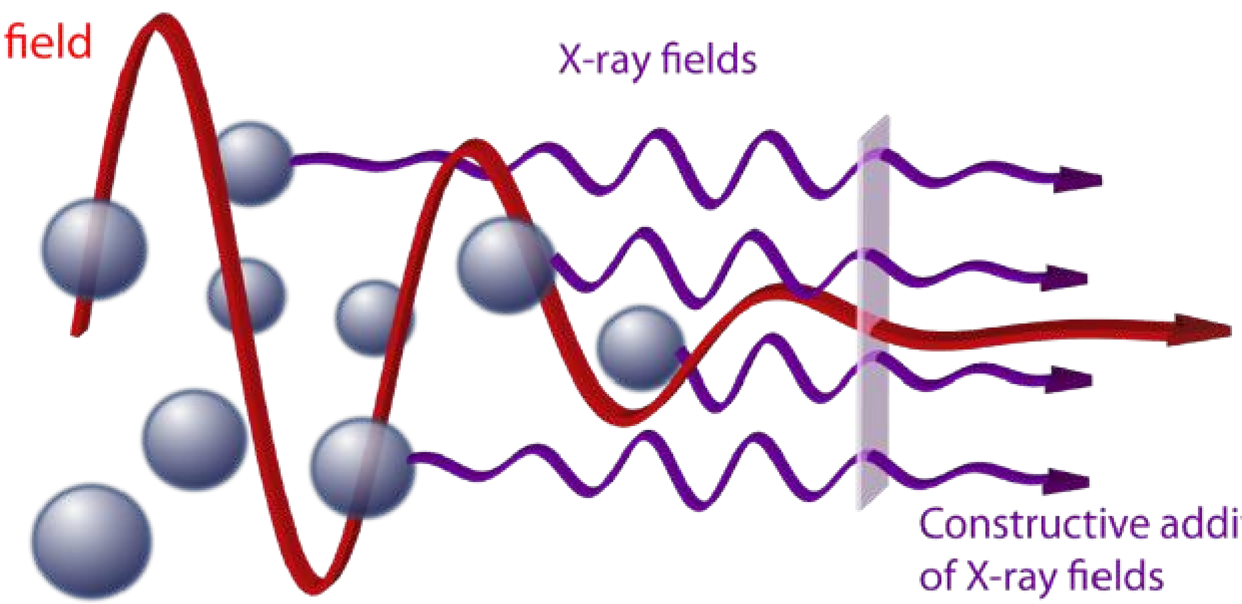
High harmonic generation (HHG)

*JOSA B **4**, 595 (1987)*
*J Phys B **21**, L31 (1988)*

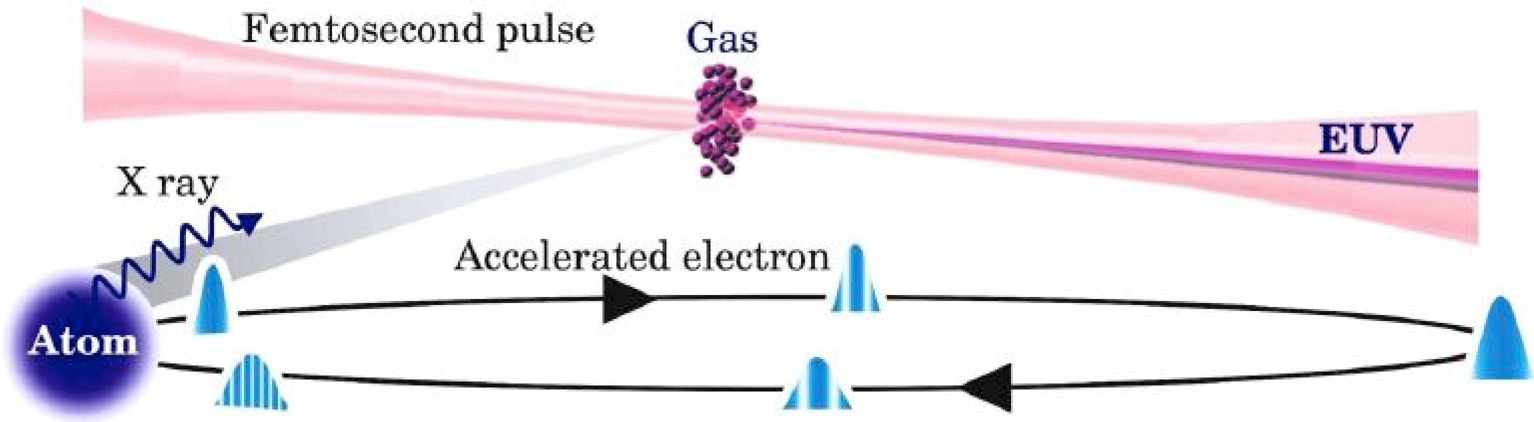


Phase matched HHG

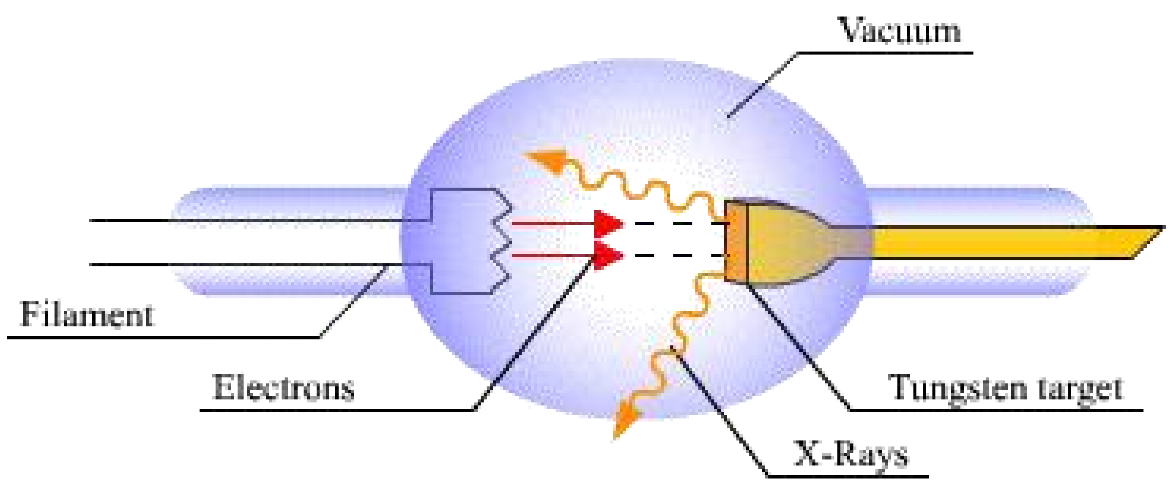
*Science **280**, 1412 (1998)*
*Science **336**, 1287 (2012)*



High harmonic generation – coherent version of the X-ray tube

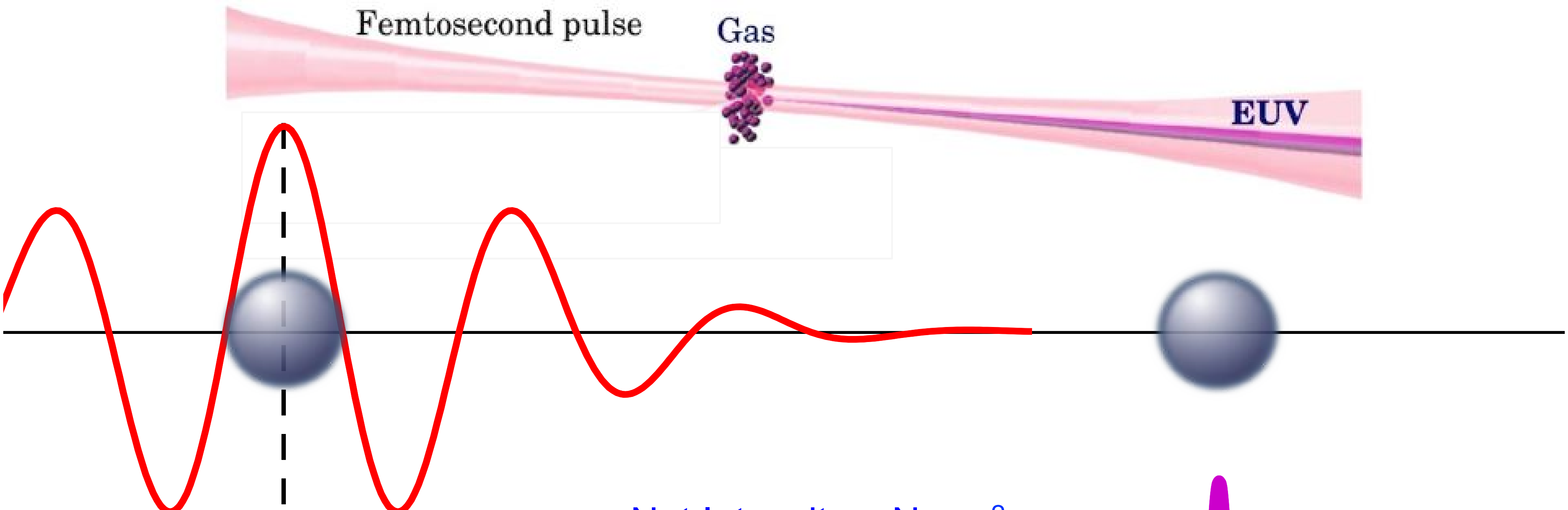


High harmonic generation *(JOSA B 4, 595 ('87); J Phys B 21, L31 ('88))*



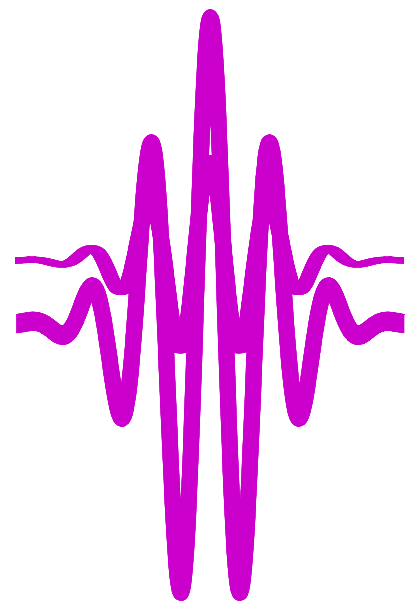
Röntgen X-ray Tube *(Roentgen, Nature (1896))*

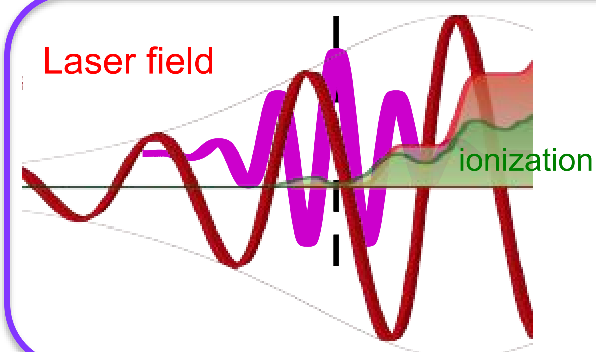
Harder challenge: “phase-matching” in plasmas



Net Intensity ~ N_{atoms}^2

PHASE MATCHED

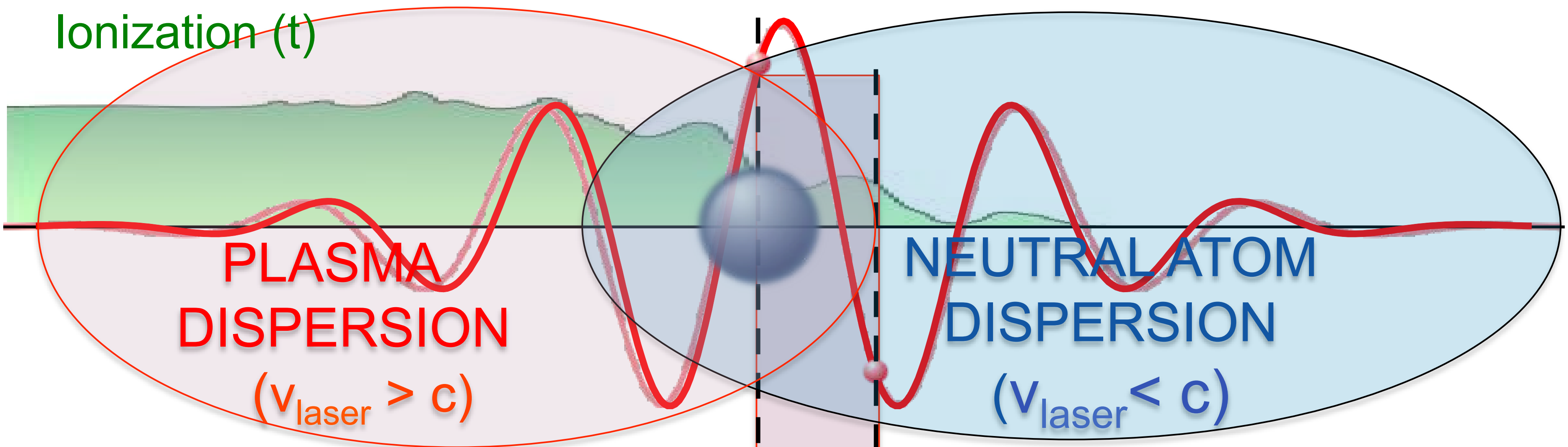


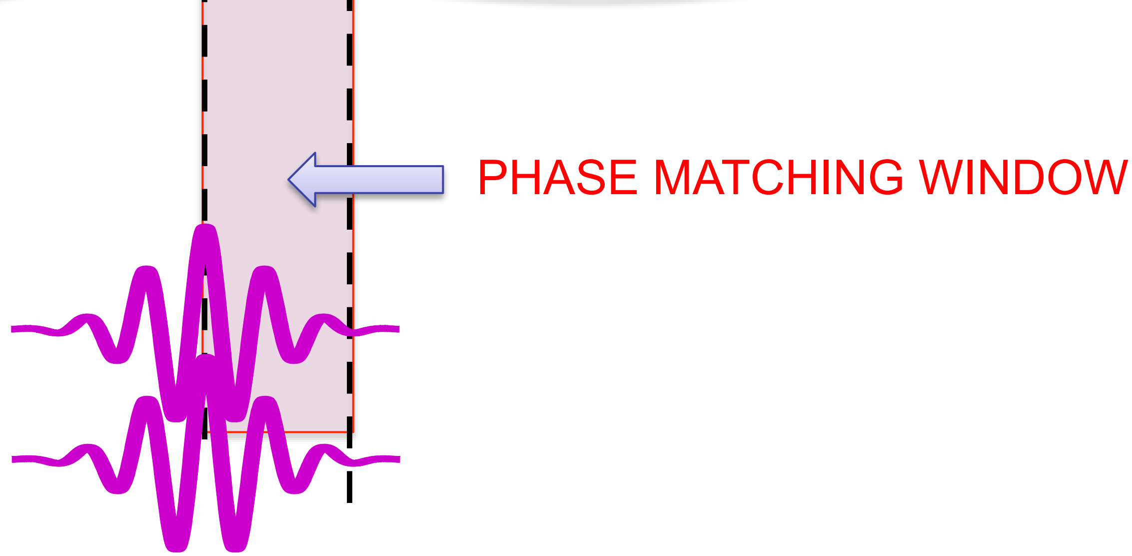


- Refractive index of laser is time dependent!
- Phase matching thought impossible!

Dynamic phase matching during small time interval

$$\Delta k = q\left\{\left(\frac{u_{11}^{2}\lambda_0}{4\pi a^{2}}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta\left[N_{atm} r_e \lambda_0\right]\right)\right\}$$

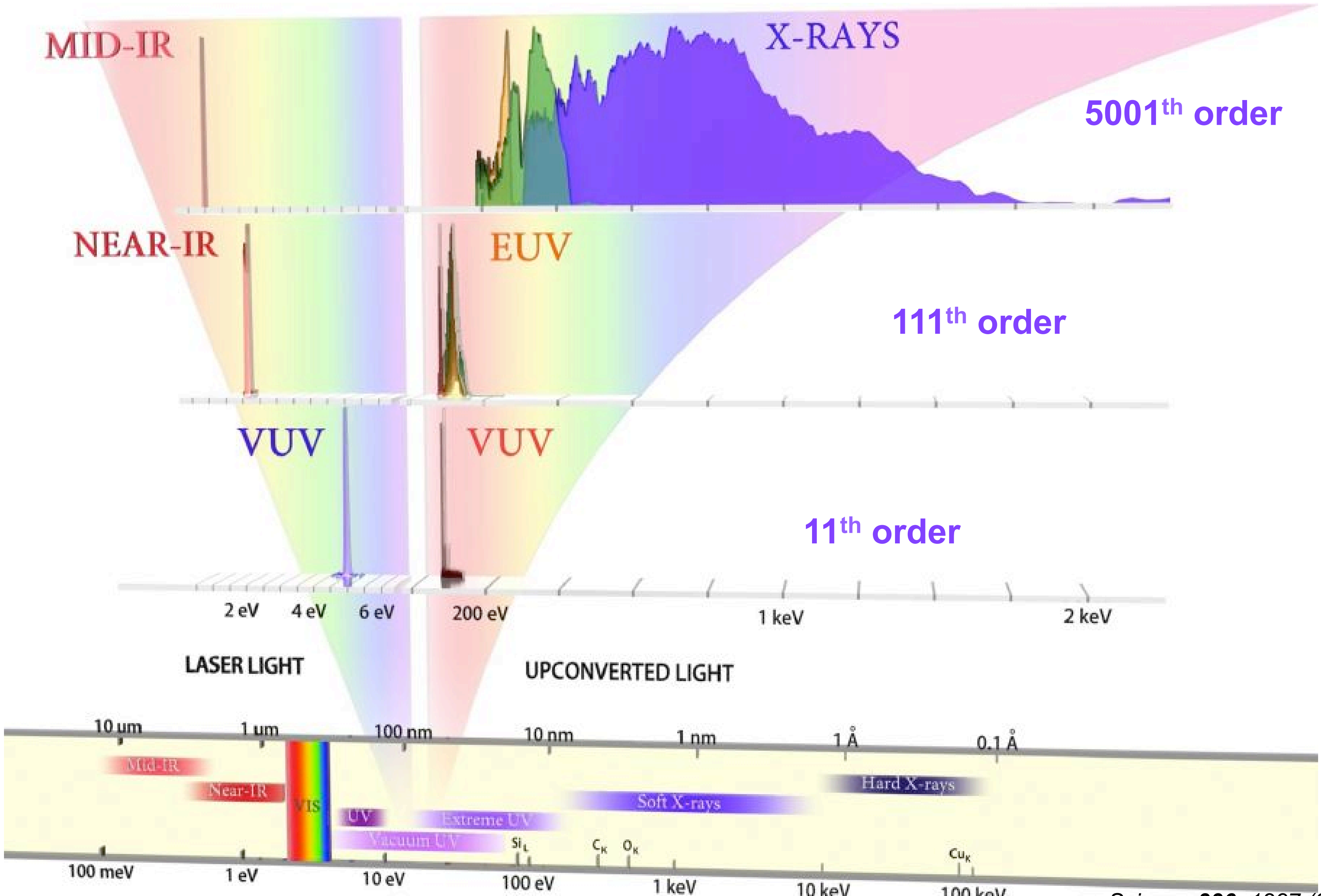




Dynamic phase matching

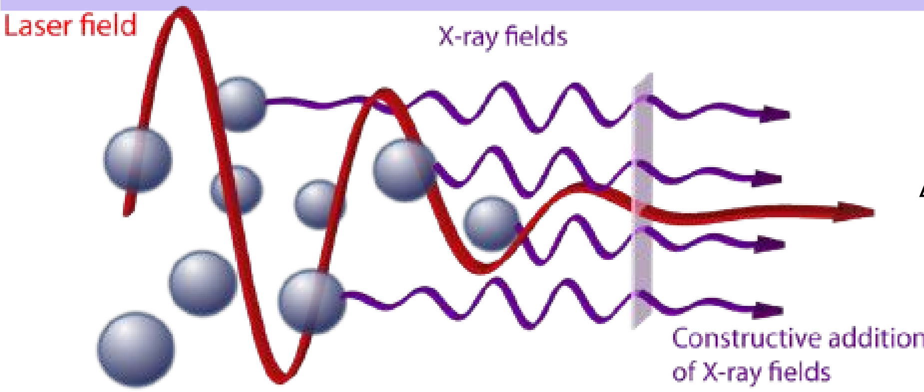
- **v_{laser} = c** during one half cycle
- Waves synchronized due to tunnel ionization and phase matching window

Counterintuitive extreme nonlinear optics



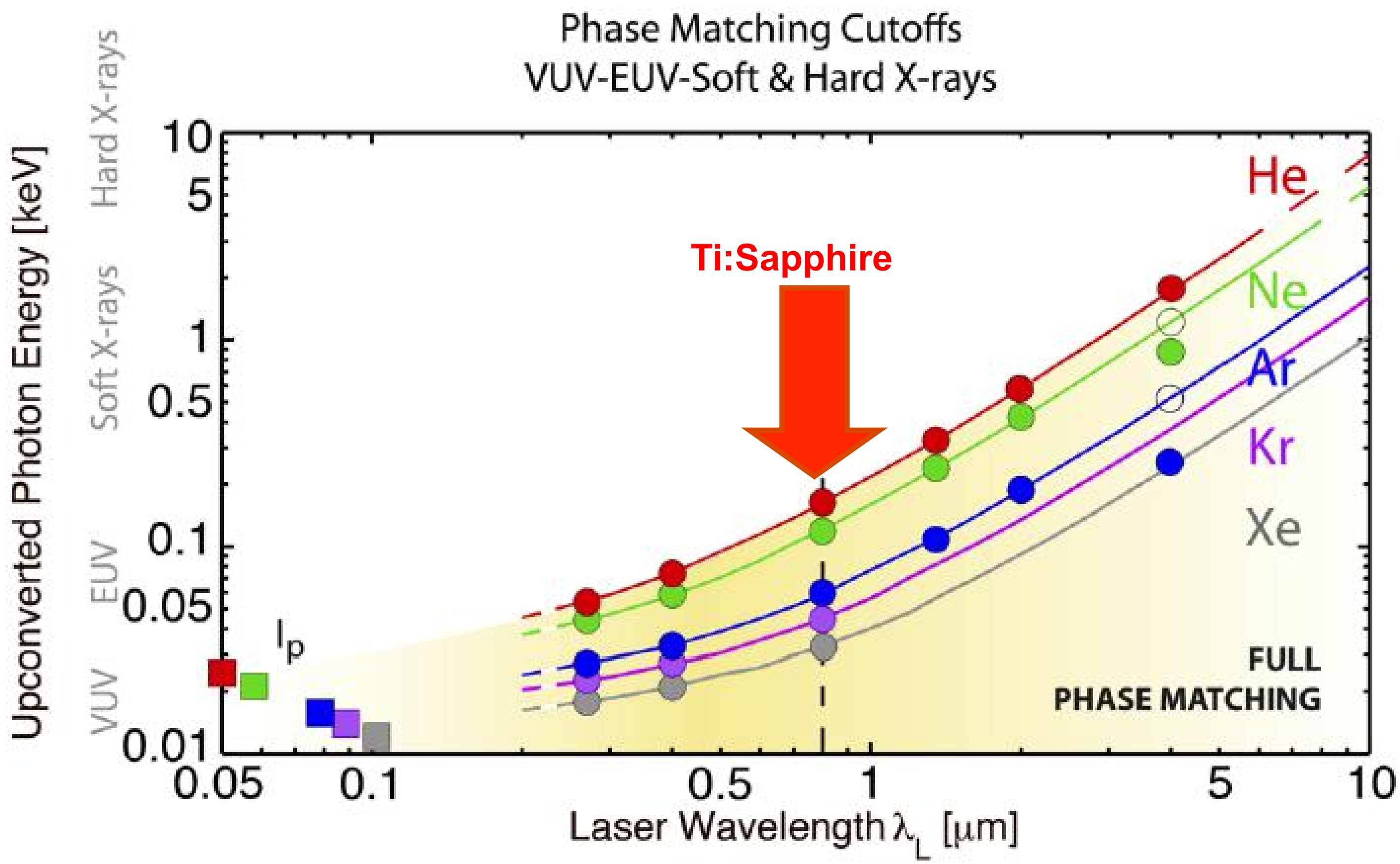
Science ***336***, *1287 (2012)*

Bright HHG emission driven by ultrafast mid-IR lasers



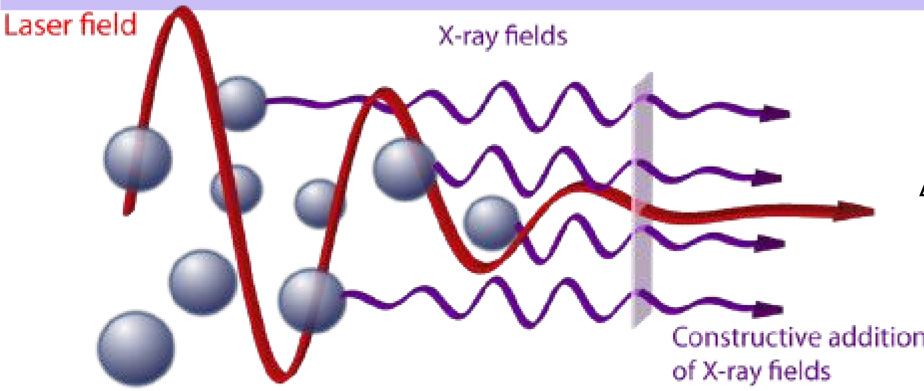
$$\Delta k = q\left\{\left(\frac{u_{11}^2\lambda_0}{4\pi a^2}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta\left[N_{atm} r_e \lambda_0\right]\right)\right\}$$

- **keV** HHG needs mid IR lasers
- **EUV** HHG needs 0.8μm lasers
- **VUV** HHG needs UV lasers



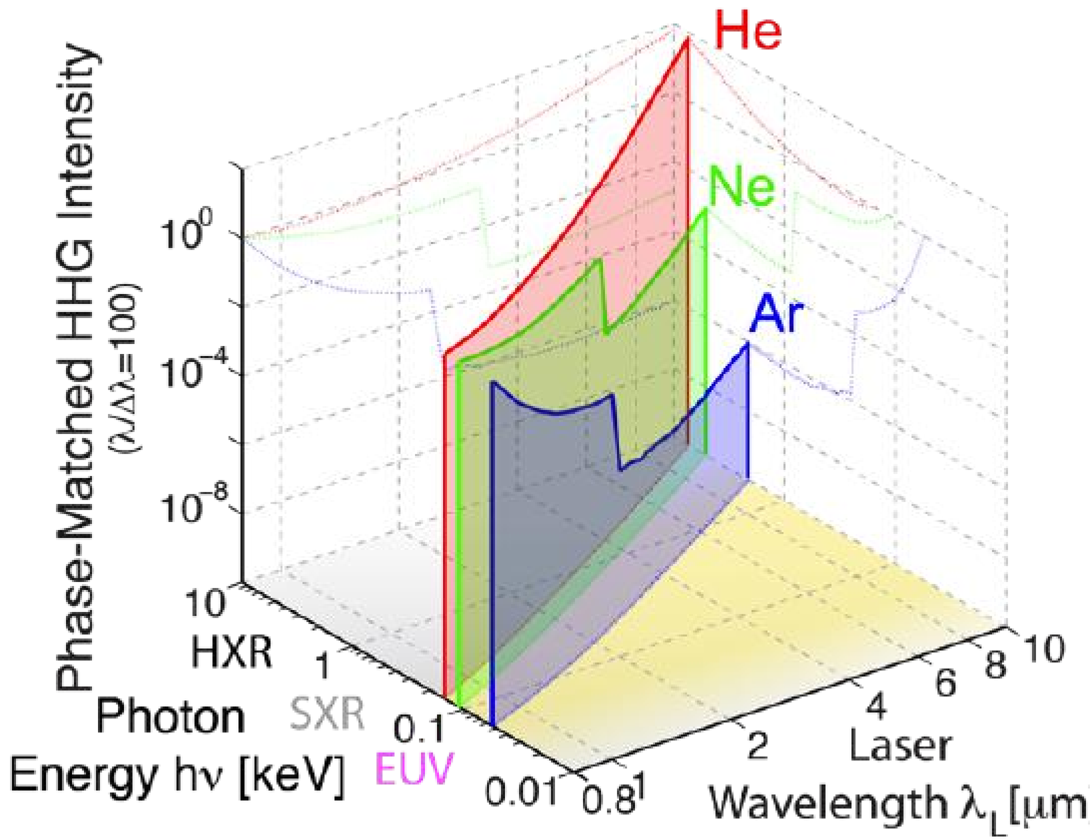
*PNAS **106**, 10516 (2009)*
*Nature Photonics **4**, 822 (2010)*
*PRL **105**, 173901 (2010)*
CLEO Postdeadline (2011)
*Science **336**, 1287 (2012)*
Pat. No. 8,462,824 (2013)

Predict phase-matched HHG yield bright from VUV to keV



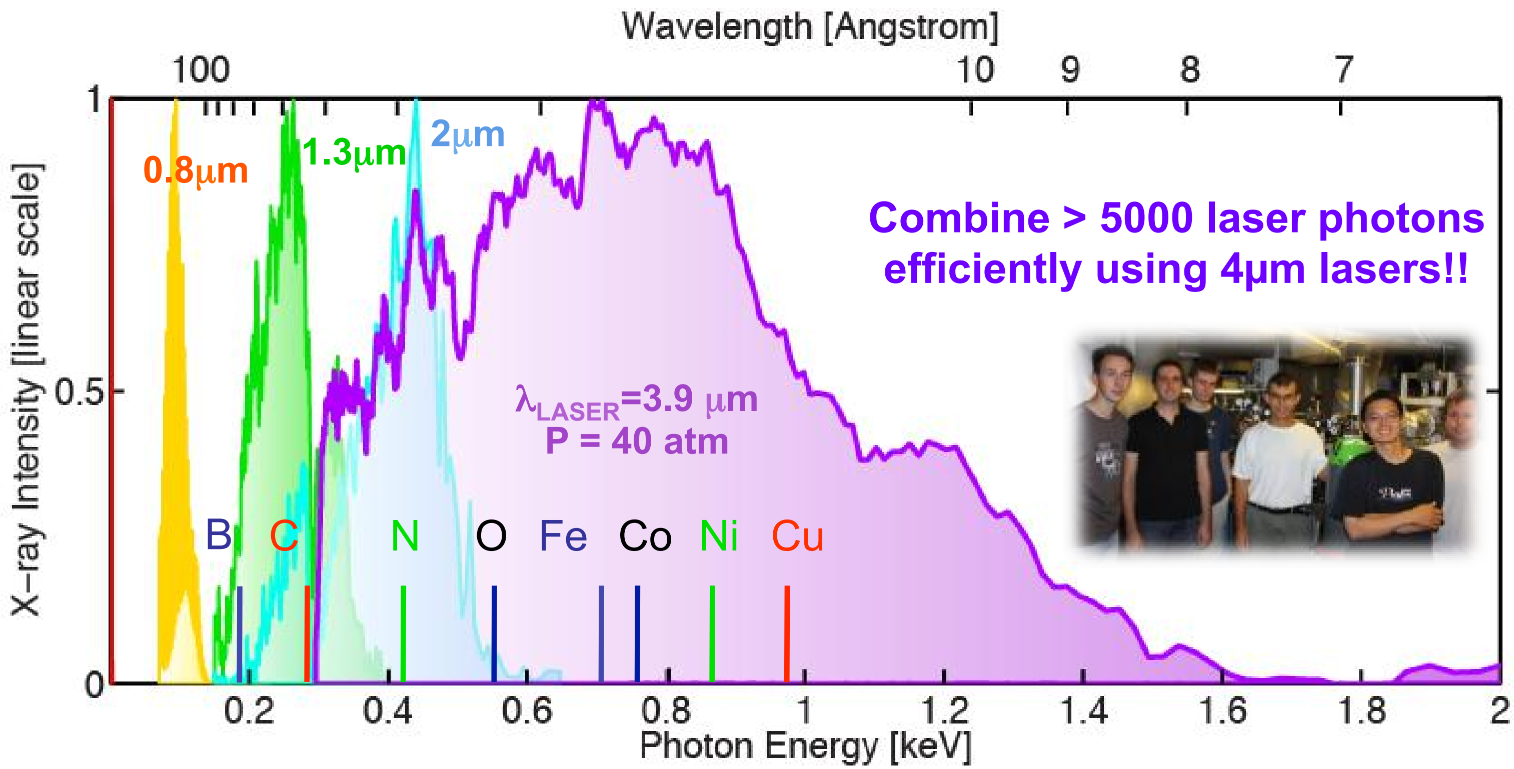
$$\Delta k = q\left\{\left(\frac{u_{11}^2\lambda_0}{4\pi a^2}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta\left[N_{atm} r_e \lambda_0\right]\right)\right\}$$

- Phase matching pressure increases from EUV to soft x-ray region
- 30 torr to 40 atm!
- Gas transparency increases
- Compensates for low single atom yield



*PNAS **106**, 10516 (2009)*
*Nature Photonics **4**, 822 (2010)*
*PRL **105**, 173901 (2010)*
CLEO Postdeadline (2011)
*Science **336**, 1287 (2012)*
Pat. No. 8,462,824 (2013)

Unique X-ray source – coherent supercontinuum to 8Å

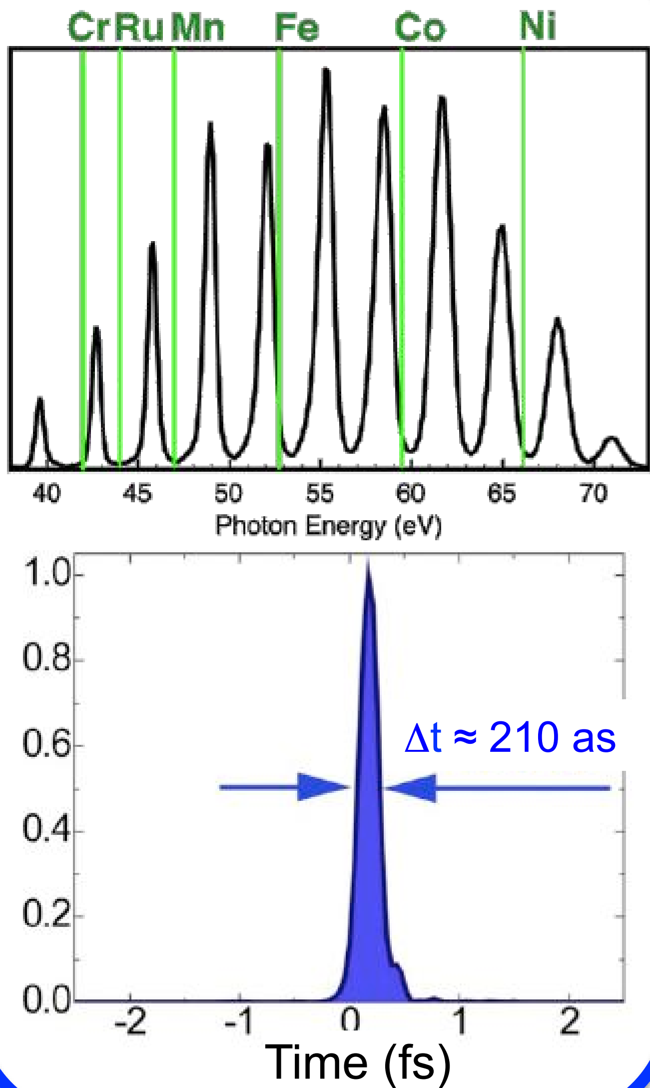


- **ONLY** bright coherent tabletop keV X-rays
- Highest nonlinear and phase matched process at > 5000 orders
- Phase matching bandwidth ultrabroad since $v_{X\text{-rays}} \approx c$
- Coherent spectrum spans many elemental x-ray edges simultaneously

*Science **336**, 1287 (2012)*

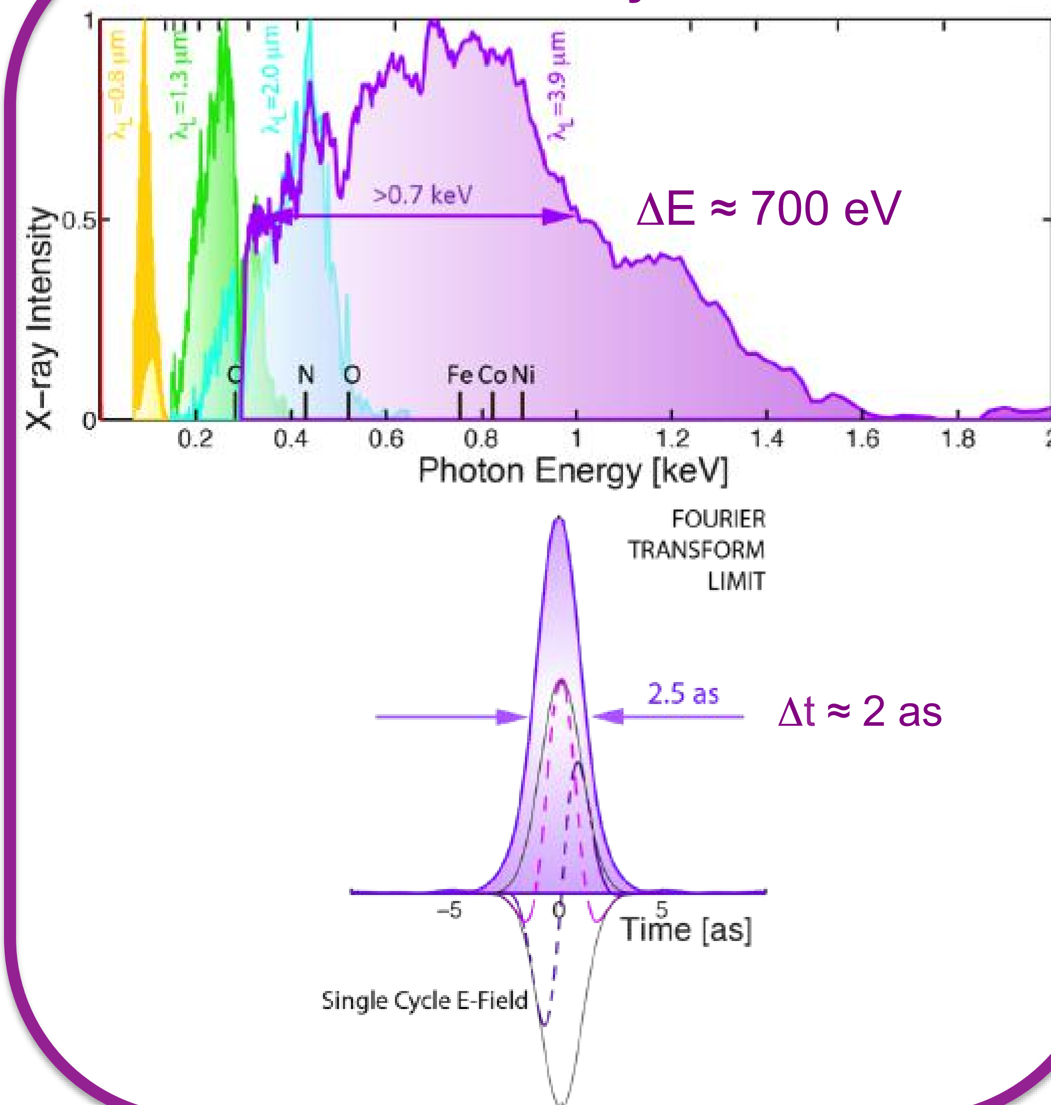
High harmonics – broad spectral coverage

EUV



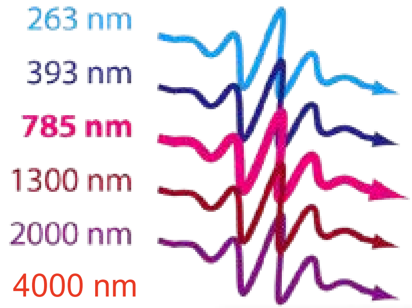
Opt. Express ***17****, 4611 (2009)*

Soft x-ray

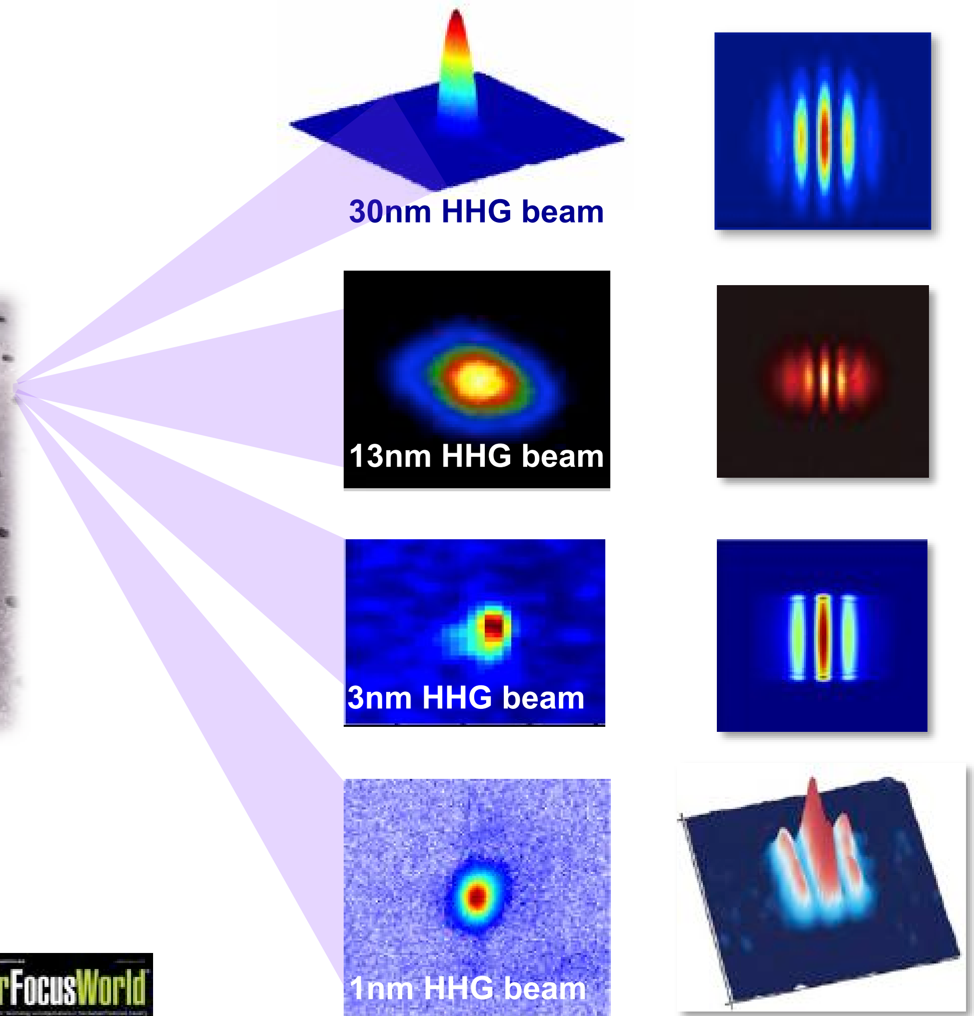


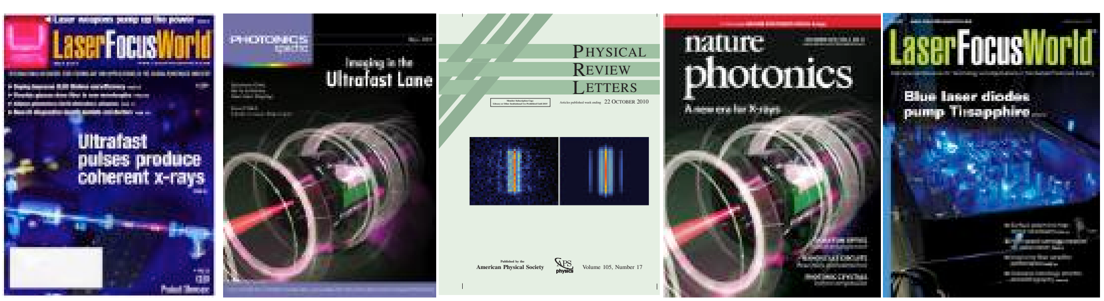
Science ***336****, 1287 (2012)*

Bright coherent beams from UV to keV



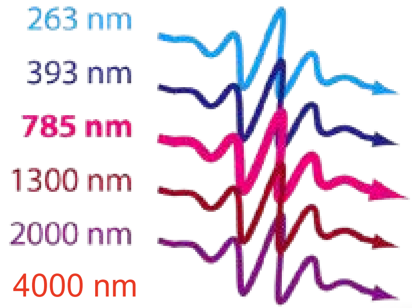
High pressure waveguide

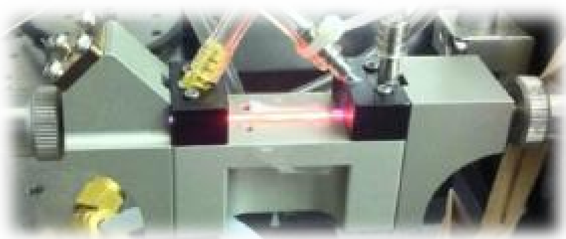


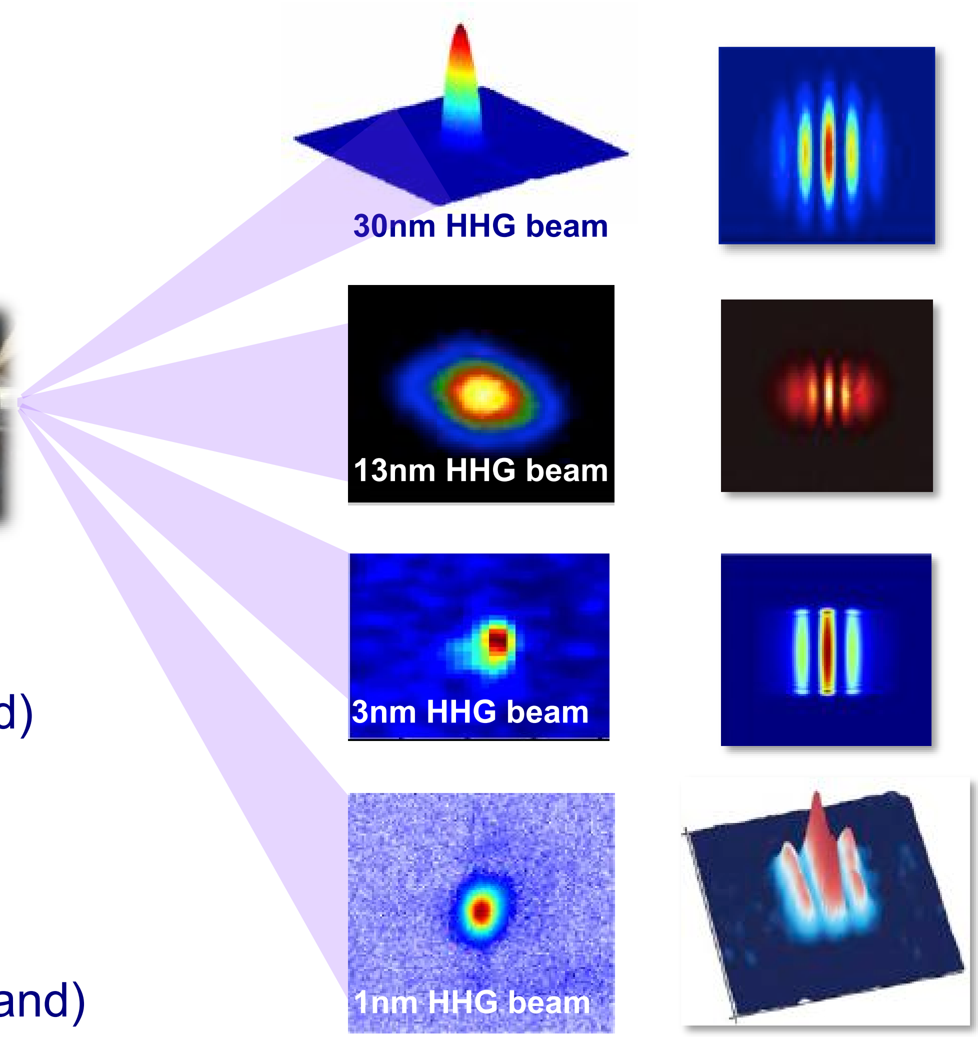


Science ***280***, *1412 (1998)*
Science ***297***, *376 (2002)*
Science ***336***, *1287 (2012)*
Pat. No. 8,462,824 (2013)

Bright coherent beams from UV to keV







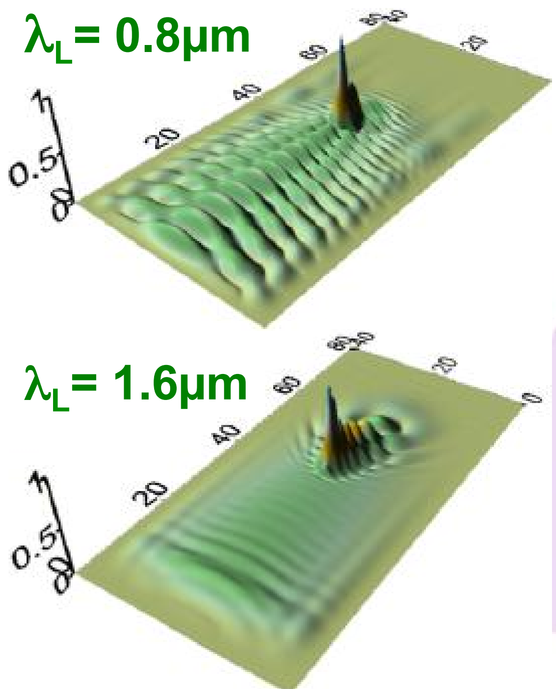
Current conversion efficiency:
10-50 eV: $10^{-3} - 10^{-4}$/eV (per 1% band)
50-100 eV: $10^{-5} - 10^{-6}$ /eV
300-1000 eV: $10^{-6} - 10^{-7}$/eV

Laser powers: 10 – 50W
EUV power: μW – 0.5mW (per 1% band)

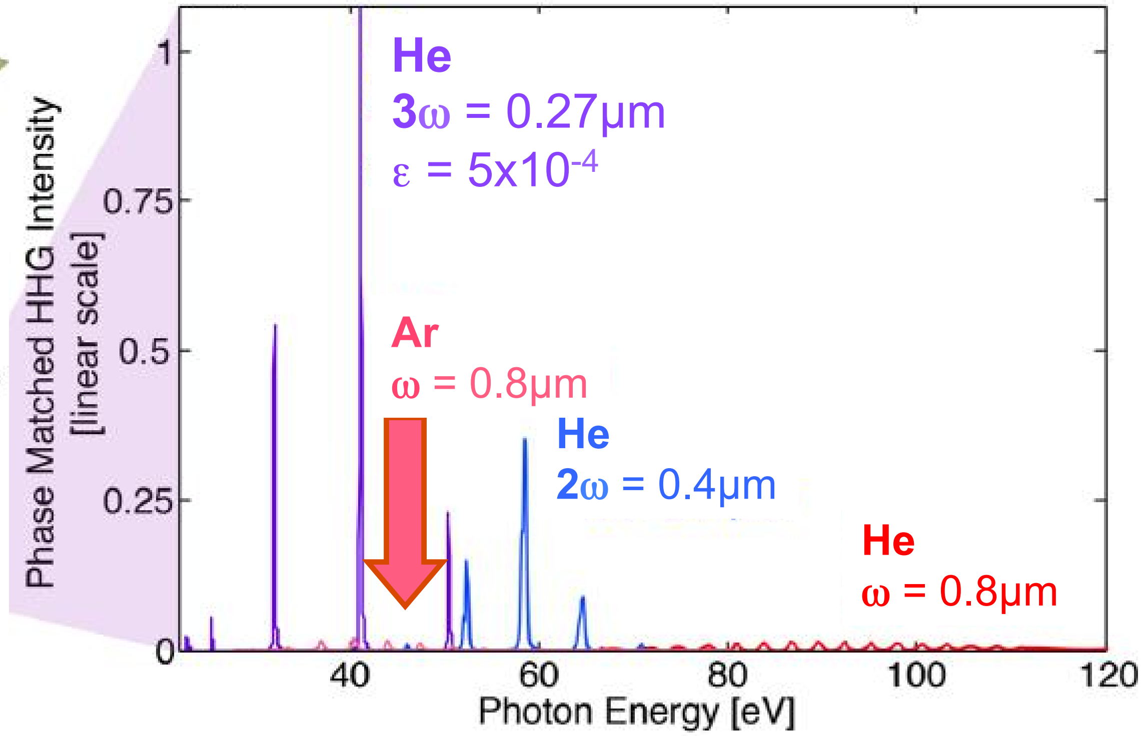
Limit not known: Increases in efficiency and photon energy very likely - new results!

Science ***280****, 1412 (1998)*
Science ***297****, 376 (2002)*
Science ***336****, 1287 (2012)*
Pat. No. 8,462,824 (2013)

Ultrahigh efficiency narrowband UV-driven harmonics



Single atom yield $\propto \lambda_L^{-5.5}$

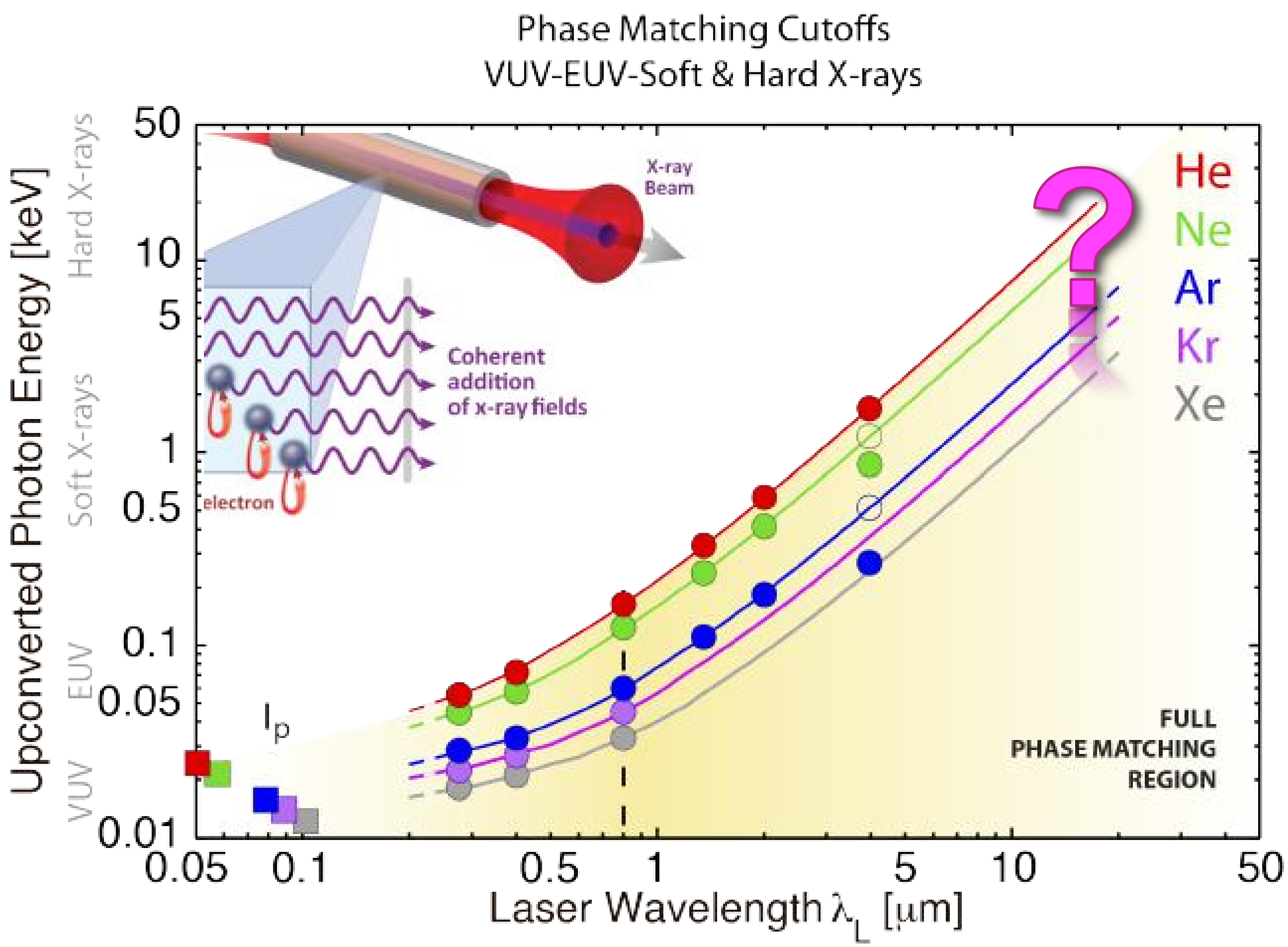


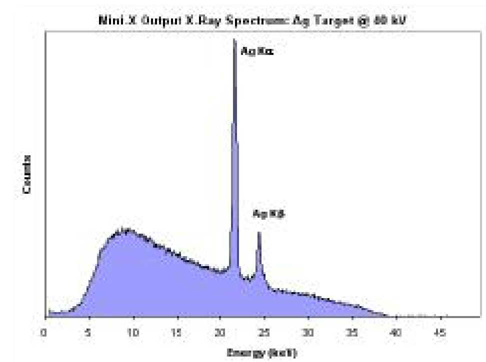
Driving HHG with 2ω and 3ω of Ti:sapphire has advantages in VUV/EUV

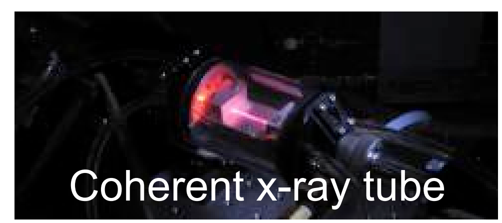
- Ultrahigh ≈ 10^{-3} efficiencies when phase matched!
- Harmonics separated by 6.2eV or 9.3eV – no need for spectral selection!
- Narrow bandwidth around 100meV – but still 10fs!
- Ideal for imaging and defect inspection at 13nm?

Limits of high harmonic generation not yet known!

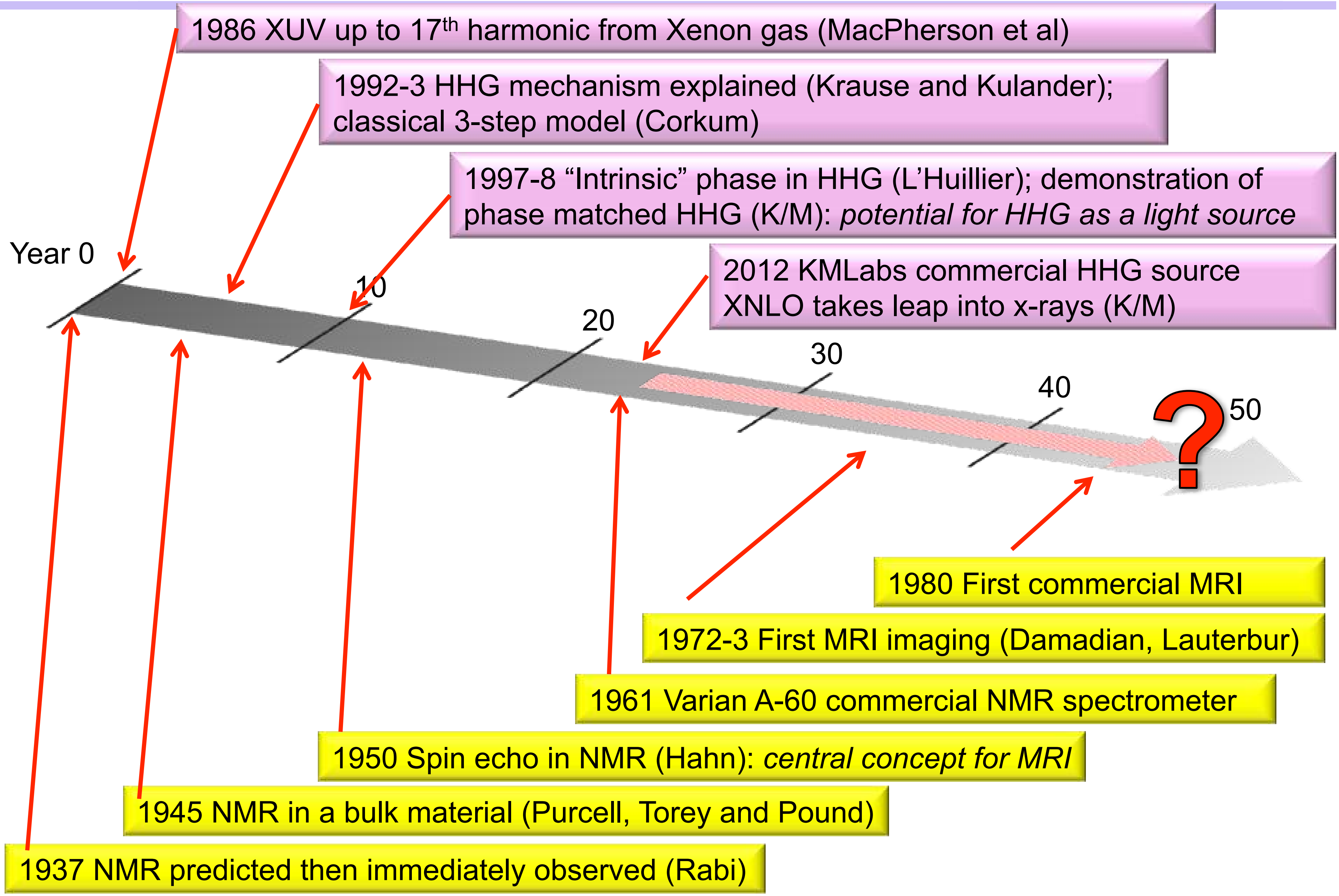
- 20 µm mid-IR lasers may generate bright 25 keV beams
- Quasi phase matching schemes also promising
- Create designer X-ray waveforms with controlled polarization state
- Potential for major disruptive technology



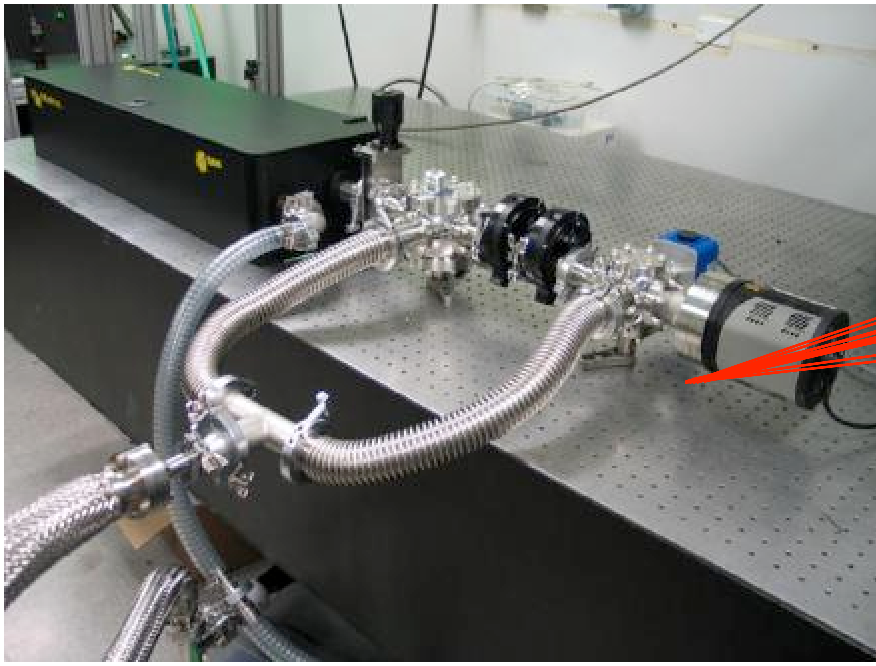


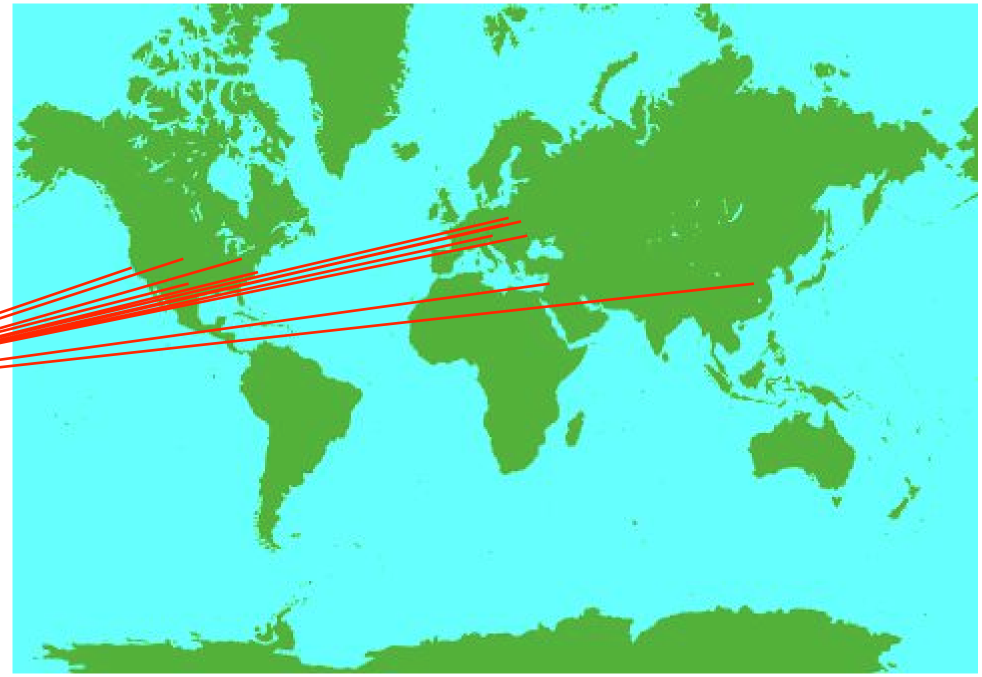


Basic phenomena ➔ broad application: 50 years



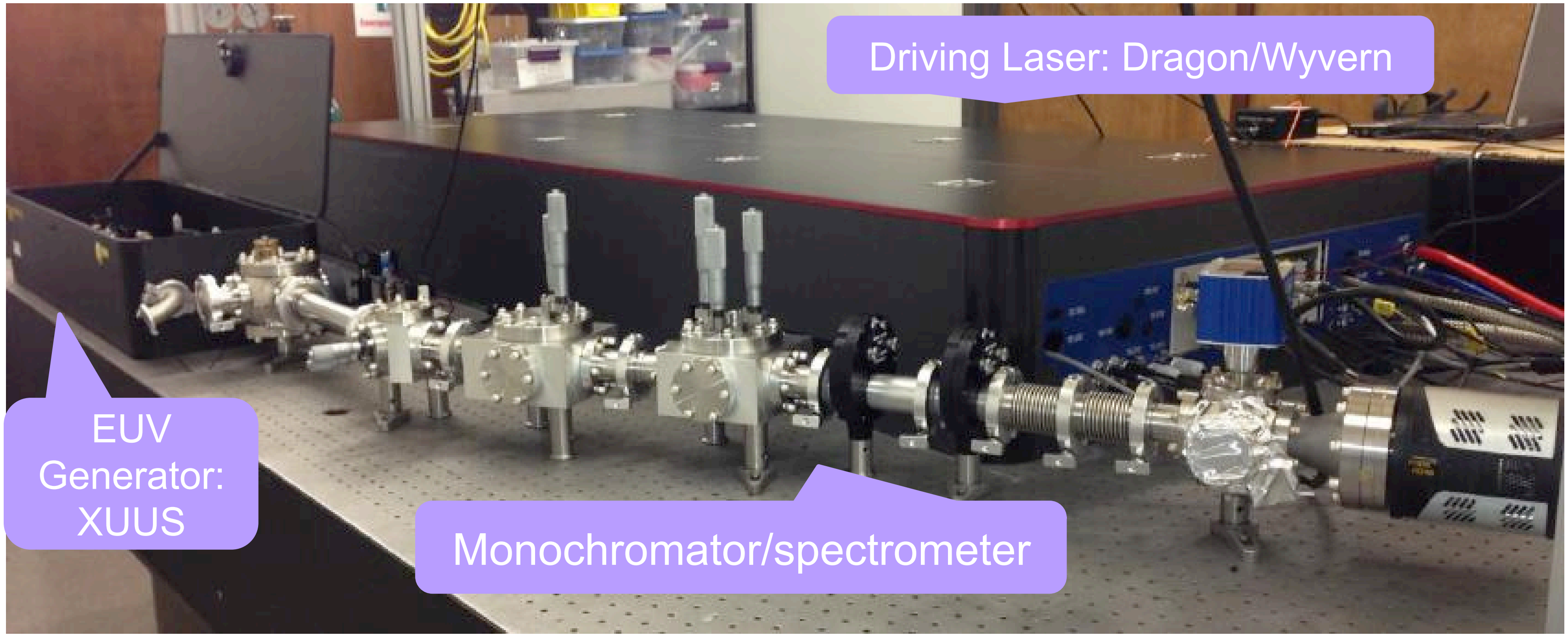
Tested in many university research labs worldwide





- First commercial ultrafast coherent EUV source for scientific market
- Operated at CLEO exhibit in May 2009
- Commercial, integrated, UHV-compatible system installed in Germany (4), Israel (2), MIT(1), Caltech (1), China (1) and Bulgaria (1) for applications in materials science
- Used successfully by many groups

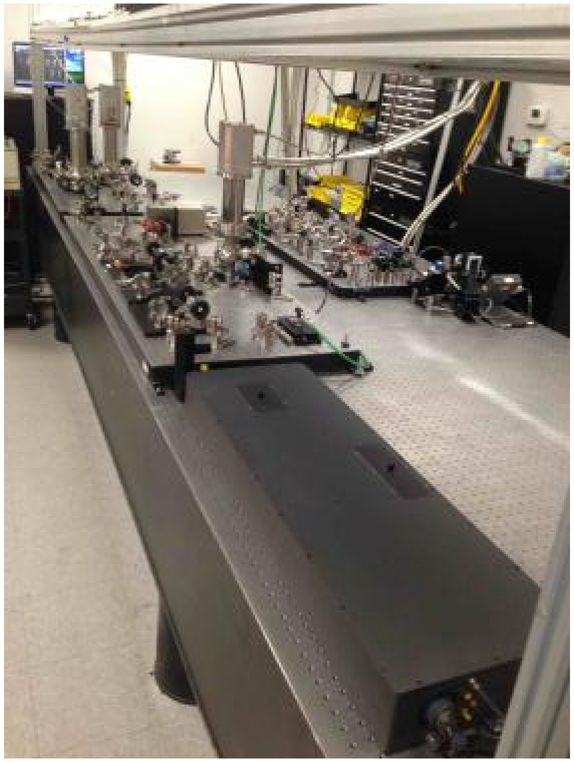
Developing robust HHG platforms



NSF 2-yr awards for 6nm and 13nm HHG
- Identify best driving laser, gas medium, waveguide designs

DOE STTR 2-yr award
- Develop compact EUV monochromator for scientific applications

Next generation lasers for 6.Xnm and beyond



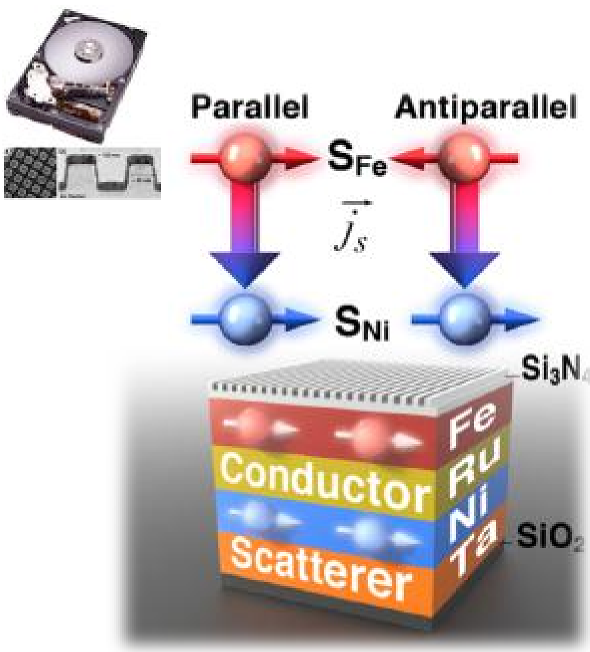
- Next generation mid-IR lasers based on chirped optical parametric amplifiers
- Robust fiber laser front end (briefcase size)
- Addresses 6.Xnm node at ≈ 190eV
- Scalable to 100kHz

DARPA 5-yr award for 1 – 6nm HHG ($7.5M between 5 groups)
- Tabletop microscope with 5nm spatial resolution

Unique nanoscience applications of EUV HHG

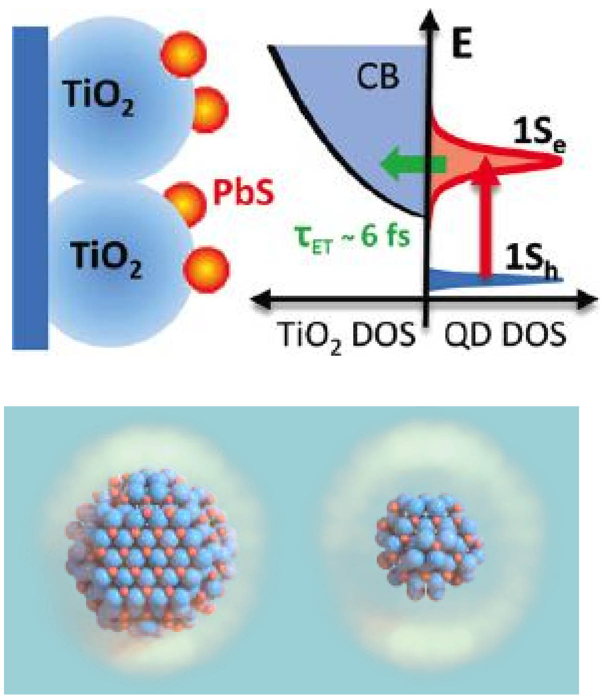
- **Understanding complex materials and nanosystems**
  - Explore correlations, many body dynamics, non-equilibrium electrons/spins, little theory
  - HHG and other new tools uncover new information and enable benchmarking with theory
  - Important technologically - data storage, nanoelectronics, energy, catalysis

Nanoscale spin dynamics



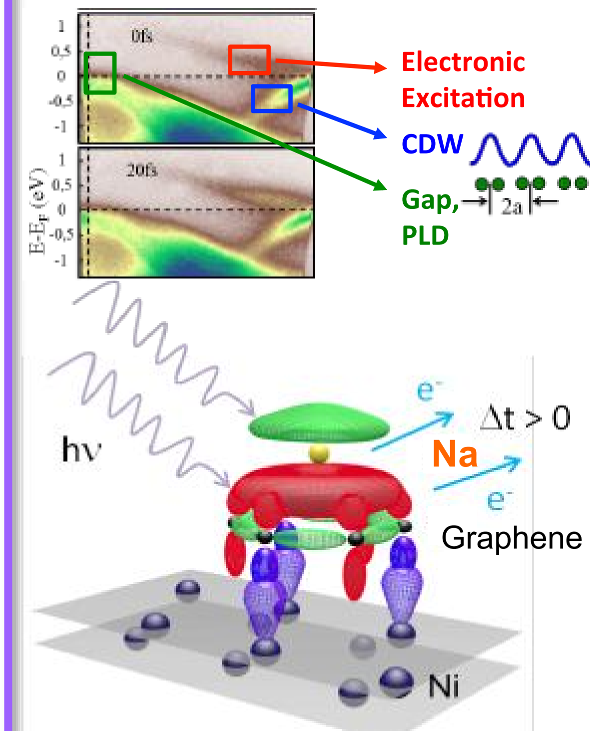
PRX ***2****, 011005 (2012); PNAS* ***109****, 4792 (2012); Nat. Commun.* ***3,*** *1037 (2012); PRL* ***110****, 197201 (2013)*

Electrons in quantum dots



Nano Letters ***13****, 2924 (2013)*
In prep (2013)

Correlated materials



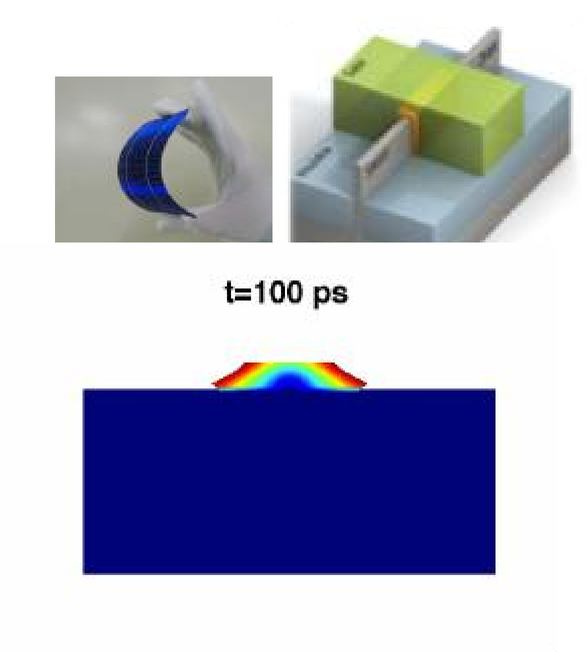
Nature ***471****, 490 (2011); Nat. Comm* ***3****, 1069 (2012); Submitted (2013)*

Unique nanoscience applications of EUV HHG

Understanding nanoscale materials requires new capabilities

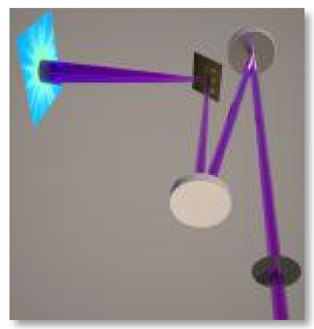
- 3D non-destructive imaging with λ spatial resolution (next generation lithography, nanoelectronics)
- Understanding nanoscale energy/charge/spin flow, no theory (thermal, strain, metamaterials)

Acoustic nanometrology



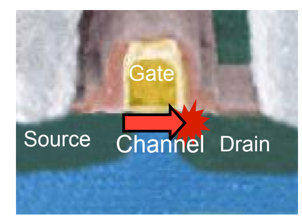
Nature Materials ***9***, *26 (2010); Nano Letters* ***11***, *4126 (2011); PRB* ***85***, *195431 (2012)*

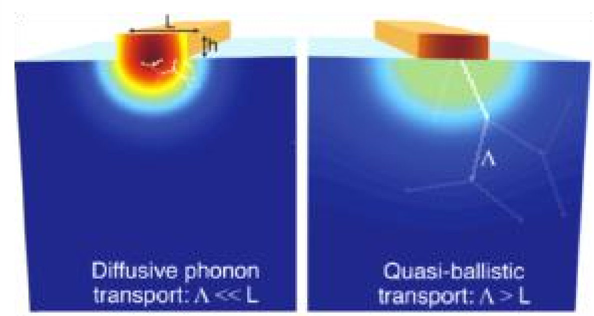
Nanoscale coherent imaging



Nature ***463***, *214 (2010); Op. Ex.* ***19***, *22470 (2011); Op. Ex.* ***17***, *19050 (2012); Opt. Ex.* ***21***, *21970 (2013) Submitted (2013)*

Nanoscale heat flow

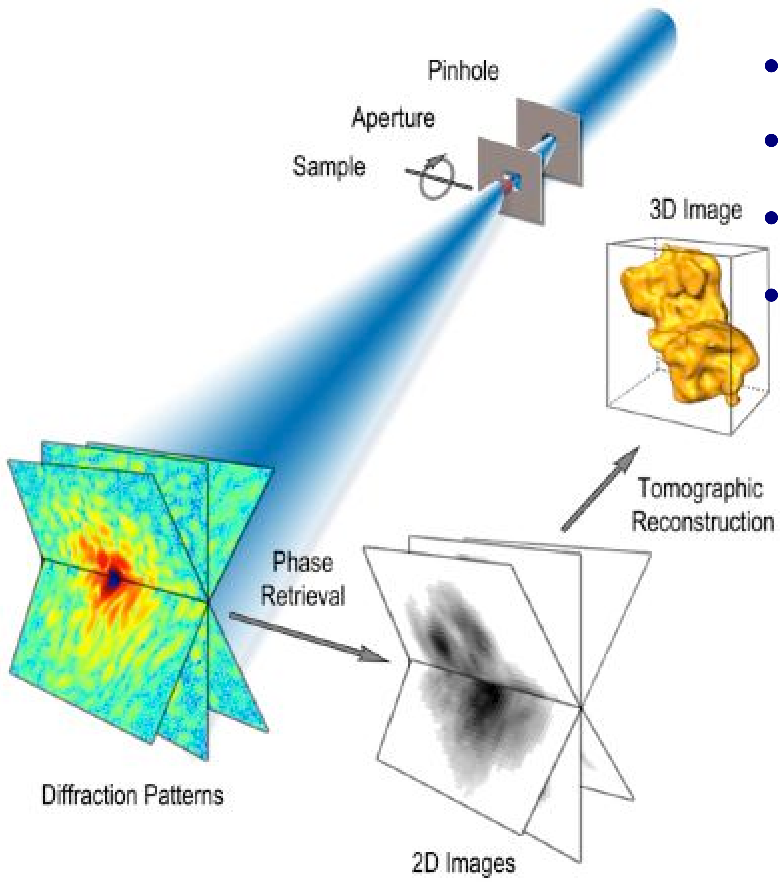


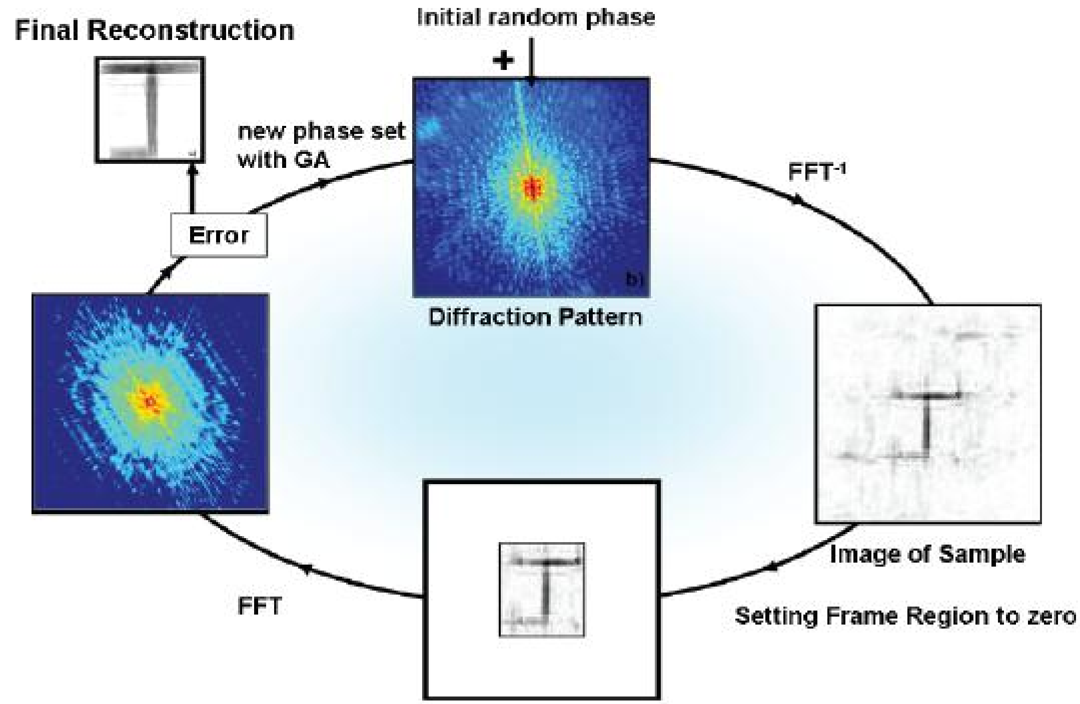


Nature Materials ***9***, *26 (2010); Nano Letters* ***11***, *4126 (2011); PRB* ***85***, *195431 (2012)*

Coherent Diffractive X-Ray Imaging (XCDI)

- **Diffraction-limited imaging ≈ $\lambda/2NA$**
- Image thick samples in 3D
- Inherent contrast for X-rays
- Robust to vibrations
- Needs a coherent beam and isolated sample

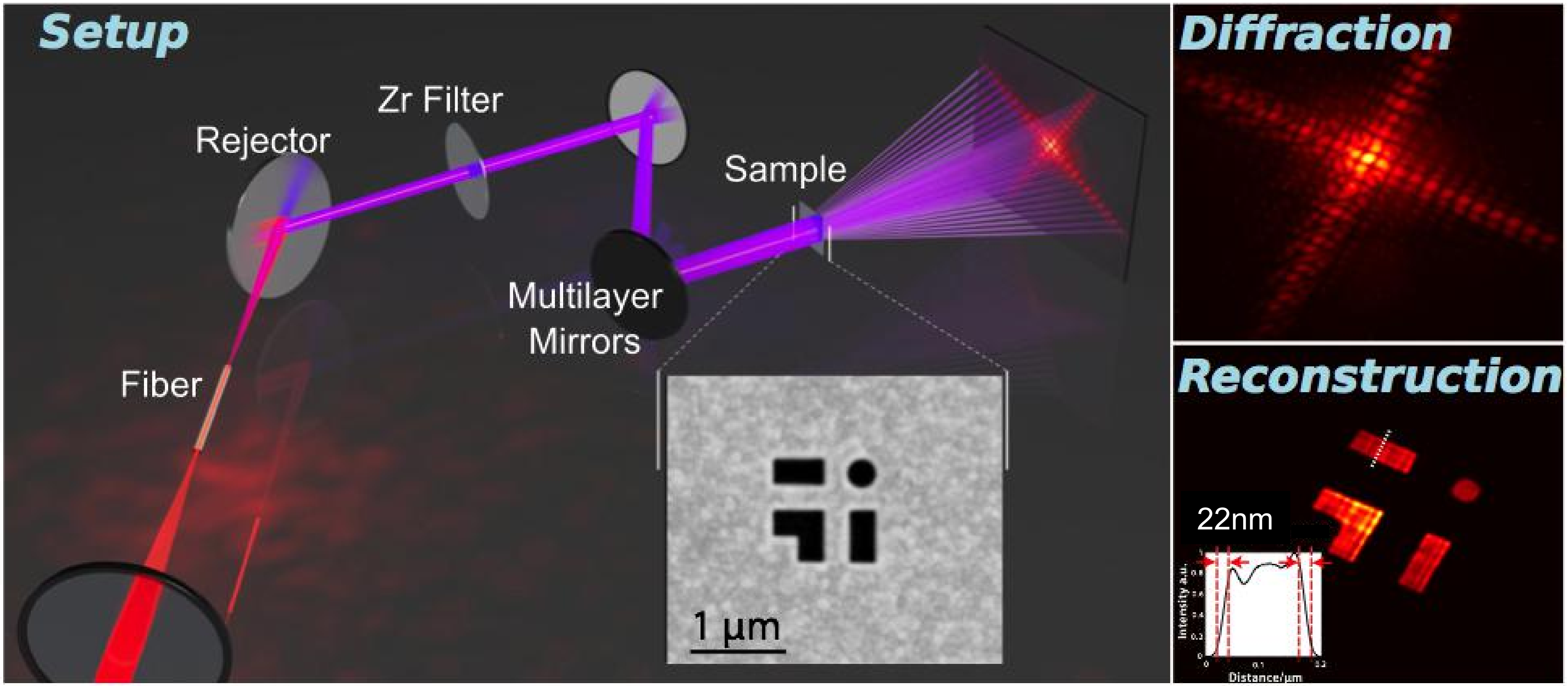




*Sayre, Acta Cryst **5**, 843 (1952)*
*Miao et al., Nature **400**, 342 (1999)*
*Miao, Nature **463**, 214 (2010)*
*Miao, Nature **483**, 444 (2012)*
*Miao, Nature **496**, 74 (2013)*

Record tabletop full field light microscope: 22nm

- NA ≈ 0.6 – 0.8
- HHG wavelength = 13 nn
- Resolution of 1.6 l or 22 nm
- BUT isolated object in transmission mode



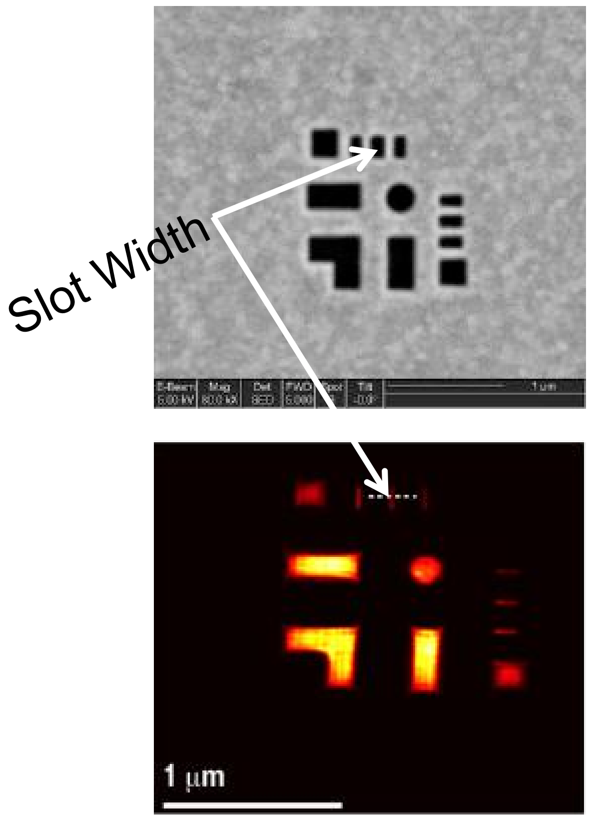
*PRL **99**, 098103 (2007); Nature **449**, 553 (2007); PNAS **105**, 24 (2008); Nature Photon. **2**, 64 (2008); OL **34**, 1618 (2009); Optics Express **19**, 22470 (2011)*

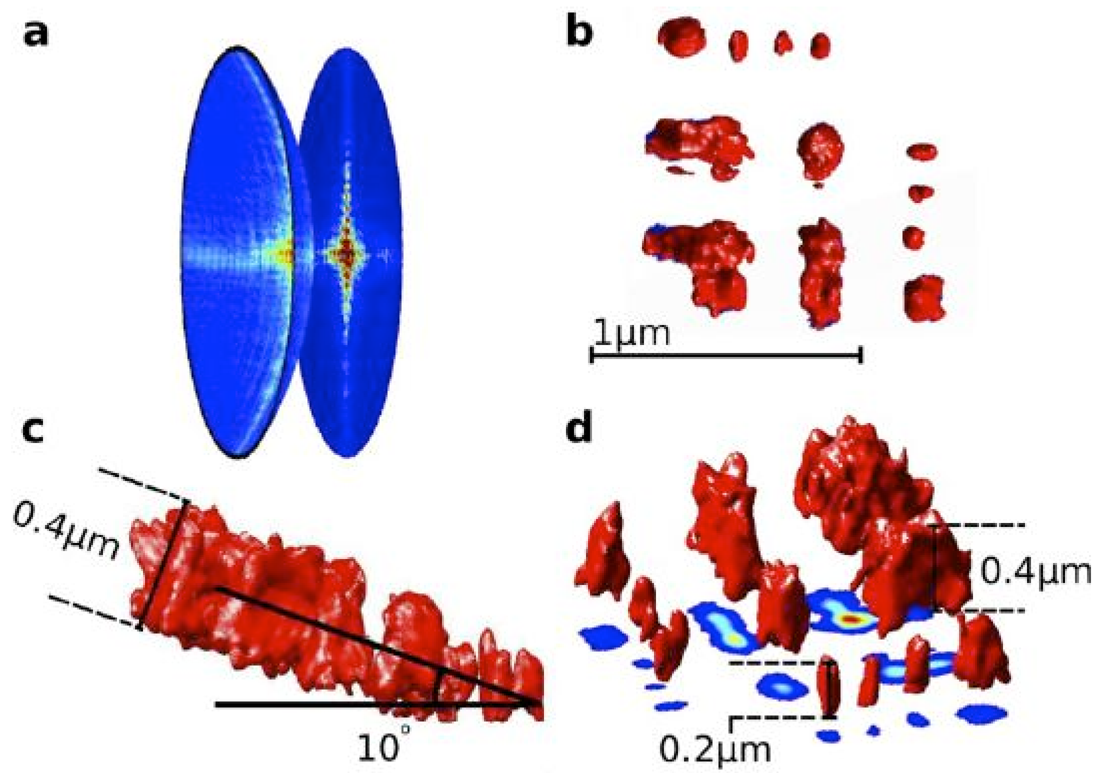
3D high numerical aperture (angle) imaging

Three-dimensional structure determination from a single view

NATURE | Vol 463 | 14 January 2010

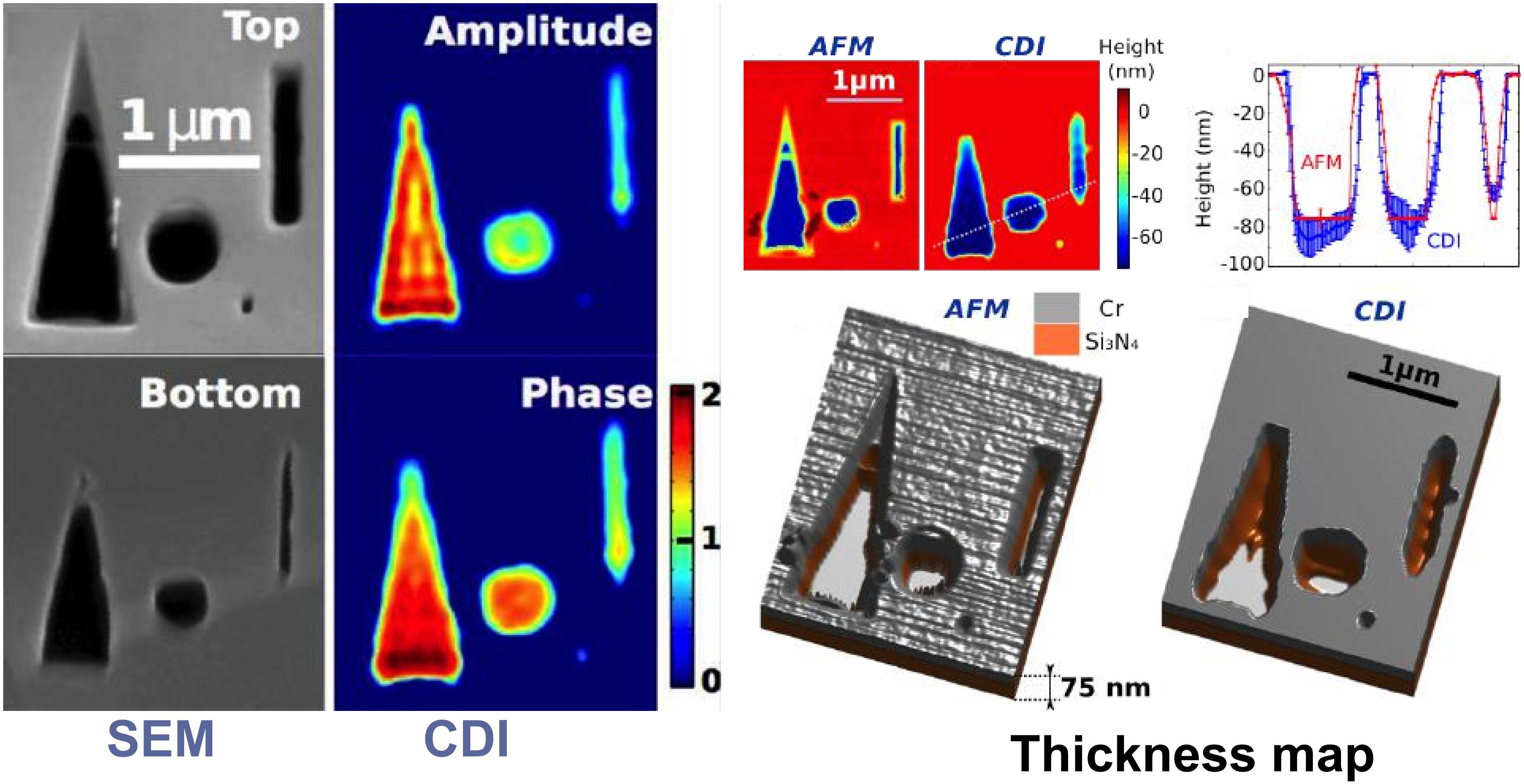
Kevin S. Raines[1,2], Sara Salha[1,2], Richard L. Sandberg[4,5], Huaidong Jiang[1,2], Jose A. Rodríguez[3], Benjamin P. Fahimian[1,2], Henry C. Kapteyn[4,5], Jincheng Du[6,7] & Jianwei Miao[1,2]





Nature ***463***, *214 (2010)*
Optics Express ***19***, *22470 (2011)*

Scanning, **non-isolated** object, transmission mode CDI

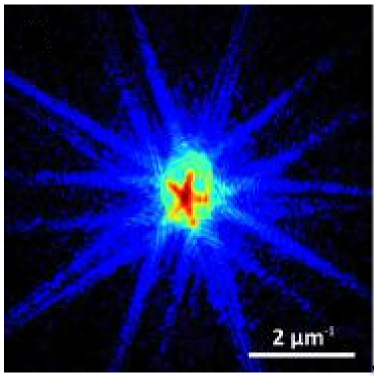


- Semi-transparent background – can extract thickness
- Non-destructive imaging compared with AFM
- 50nm hole not completely drilled through: 48nm (CDI) vs 52nm (AFM)

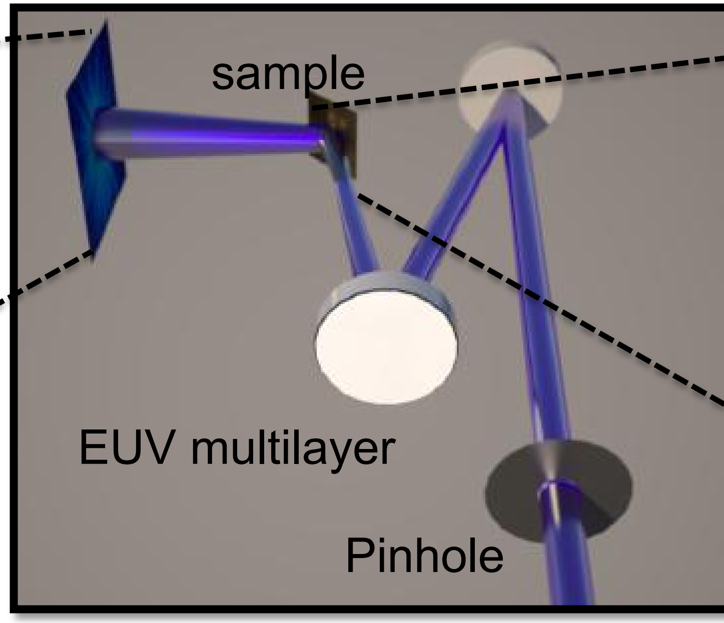
Opt. Express ***21***, *21970 (2013)*

First general, scanning, reflection mode, non-isolated object, coherent imaging on a tabletop (30nm light)

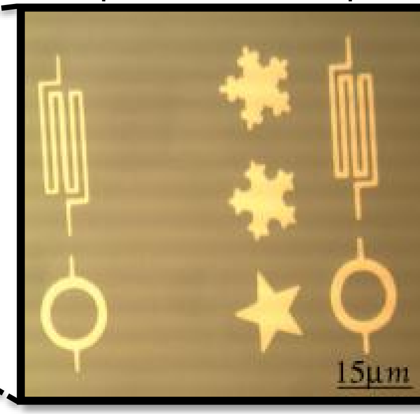
Raw CCD Data

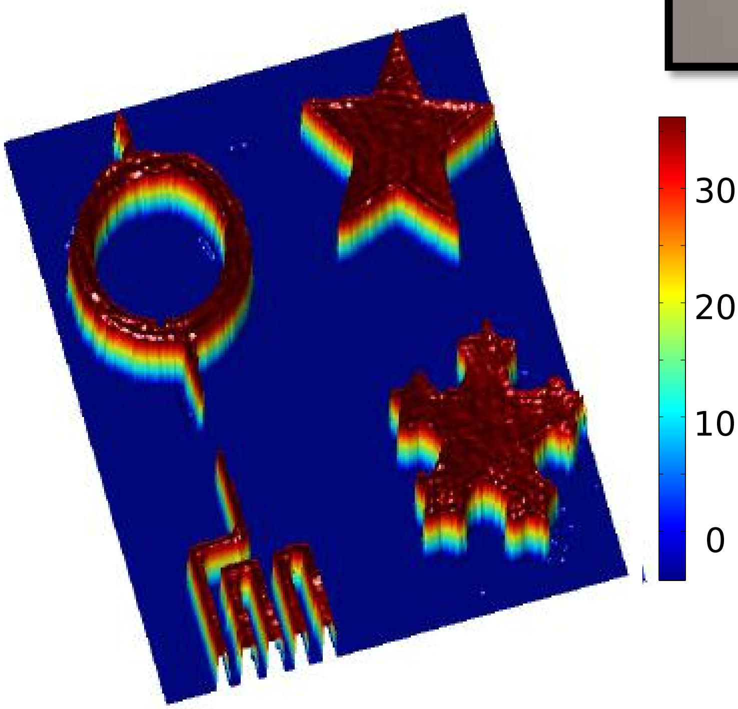


High Resolution 'Shadowgram'



Optical Microscope





EUV CDI height map in nm

- Ptychographic reconstruction recovers 31nm object height (≈ 1nm precision)
- Spatial resolution limited by NA and 30nm wavelength in this preliminary work
- Next steps: increase spatial resolution to 2λ
- Increase spatial resolution to ≈ 30nm using 13nm harmonics

First general, scanning, reflection mode, non-isolated object, coherent imaging on a tabletop (30nm light)

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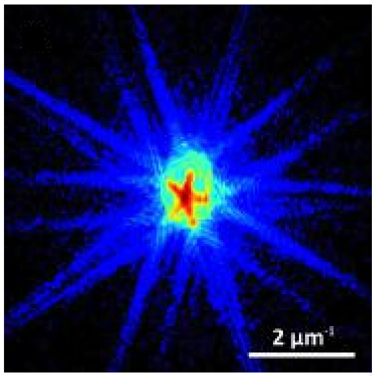

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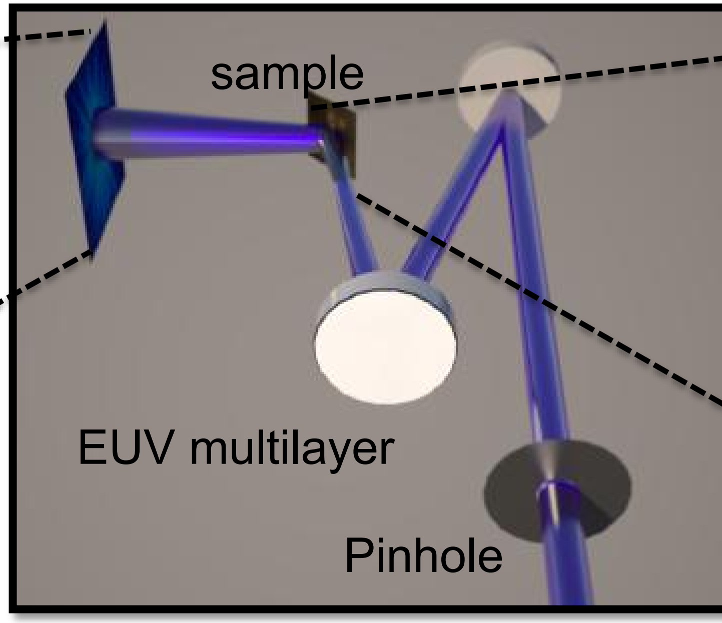
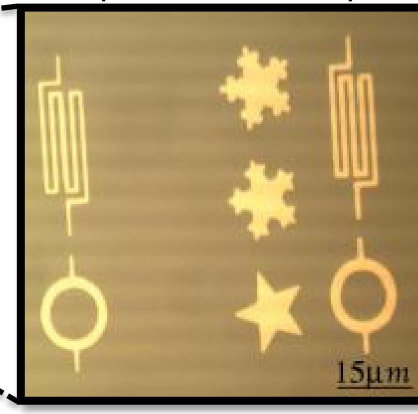
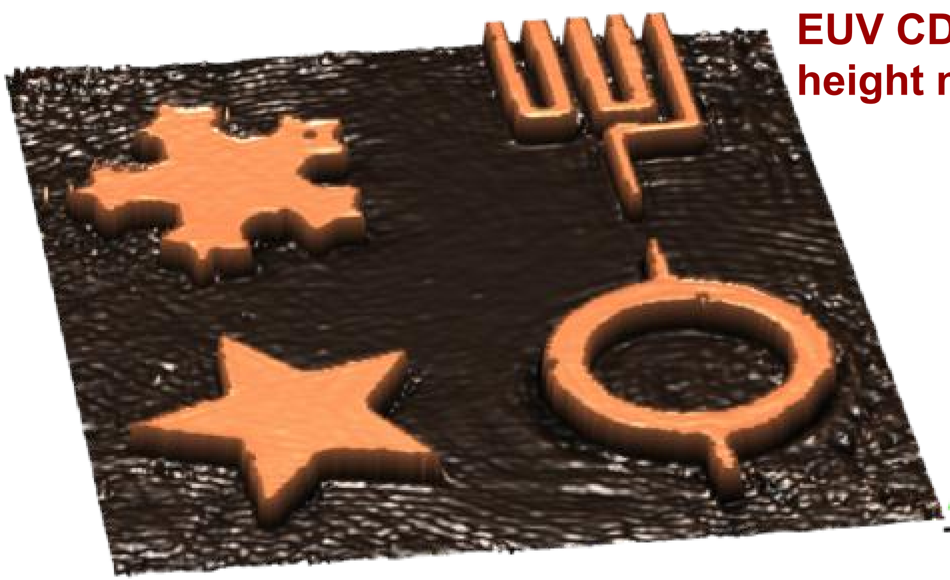


Optical Microscope





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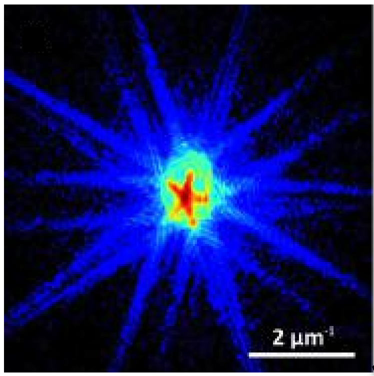

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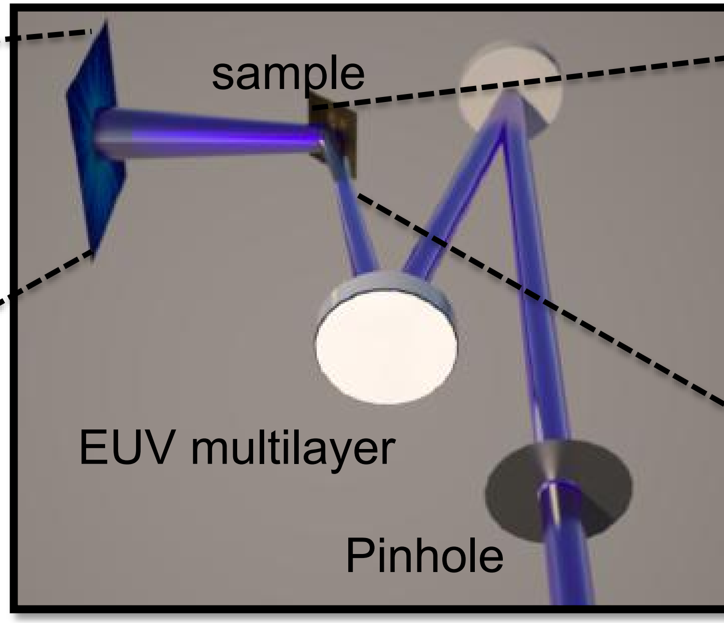

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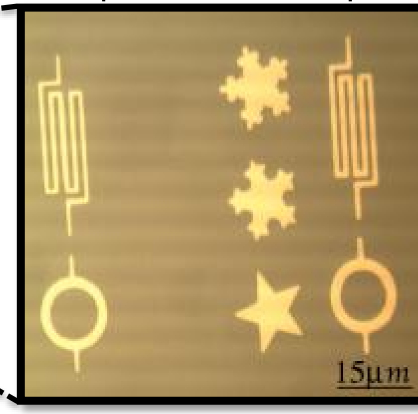
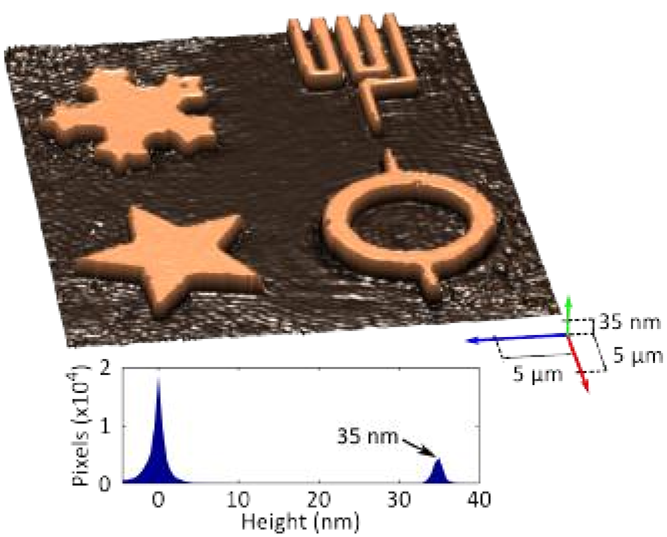
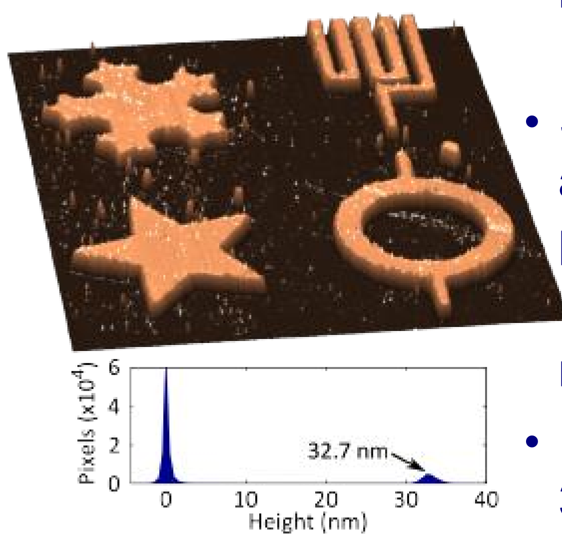


HHG CDI



AFM image



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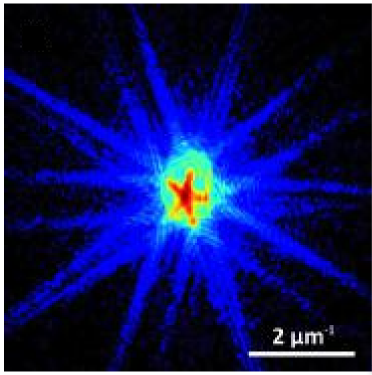
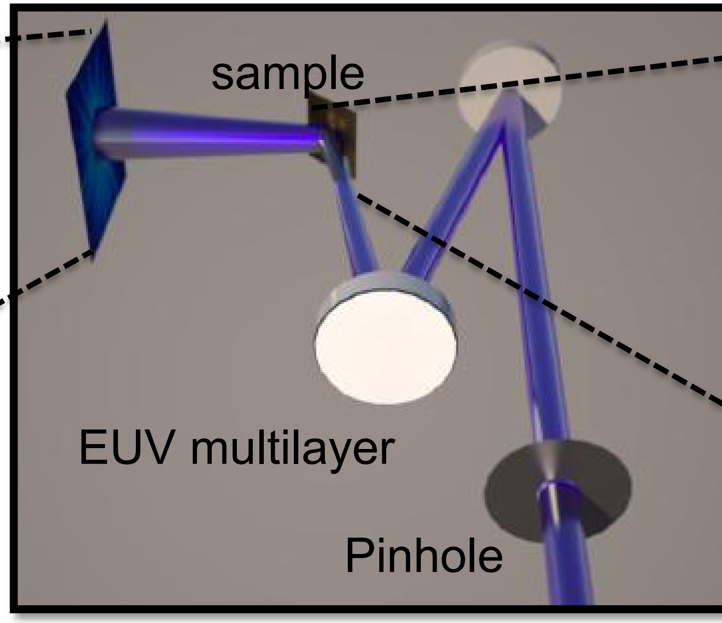


High Resolution
'Shadowgram'



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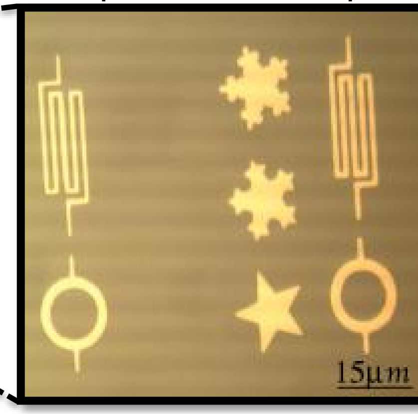
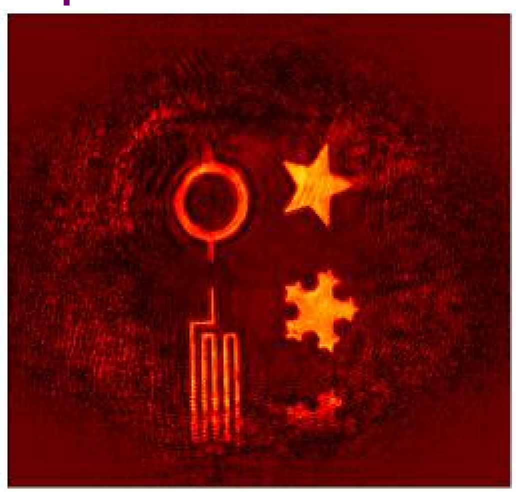


HHG CDI with position correction

HHG CDI without position correction



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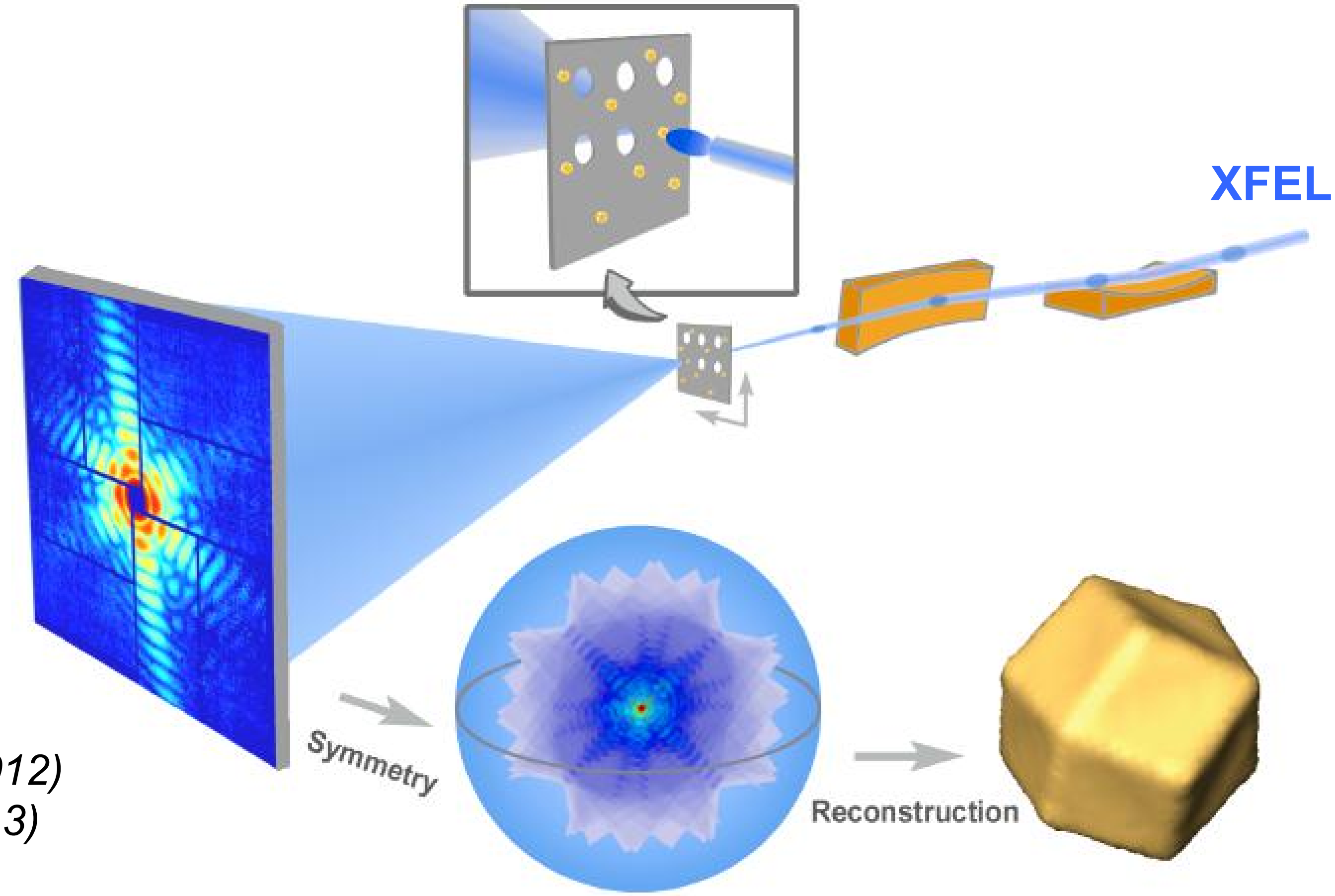
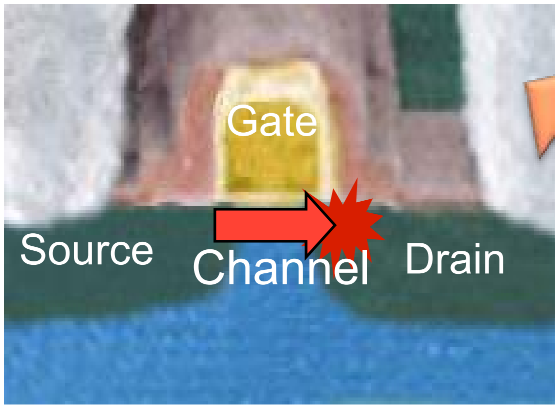
Dramatic XCDI advances using XFEL

- Single-shot 3D structure determination using femtosecond XFEL pulses at 5.4 keV (SACLA, Japan)
- 5.5nm spatial resolution, ≈ 10fs time resolution



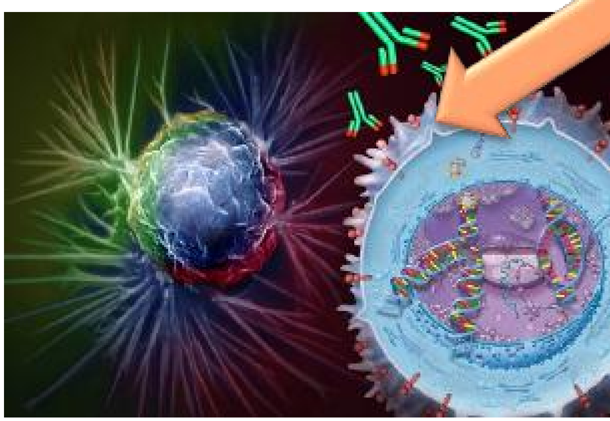
*Miao, Nature **483**, 444 (2012)*
*Miao, Nature **496**, 74 (2013)*
Miao, submitted (2013)

Importance of nano-to-bulk heat transfer

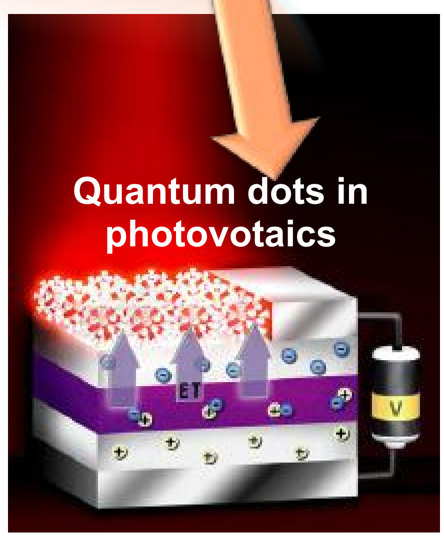


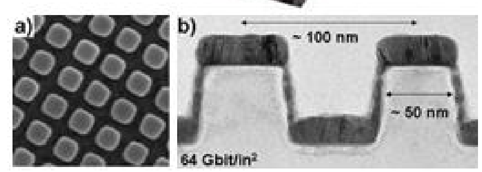
MOSFET(IBM)

TE generator (BMW)



Nano-thermal therapy for cancer





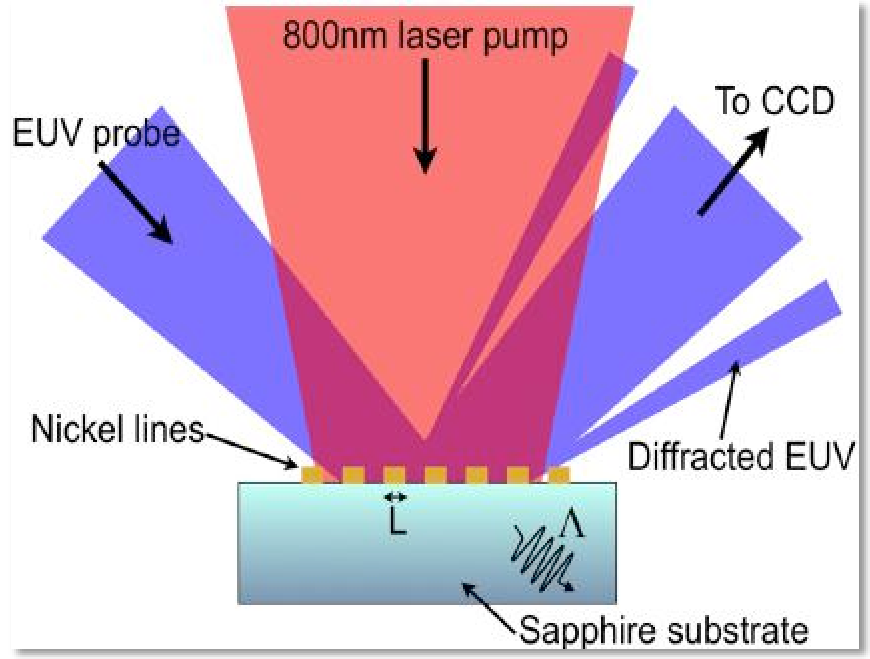
Nano patterned hard
drive (Hitachi)

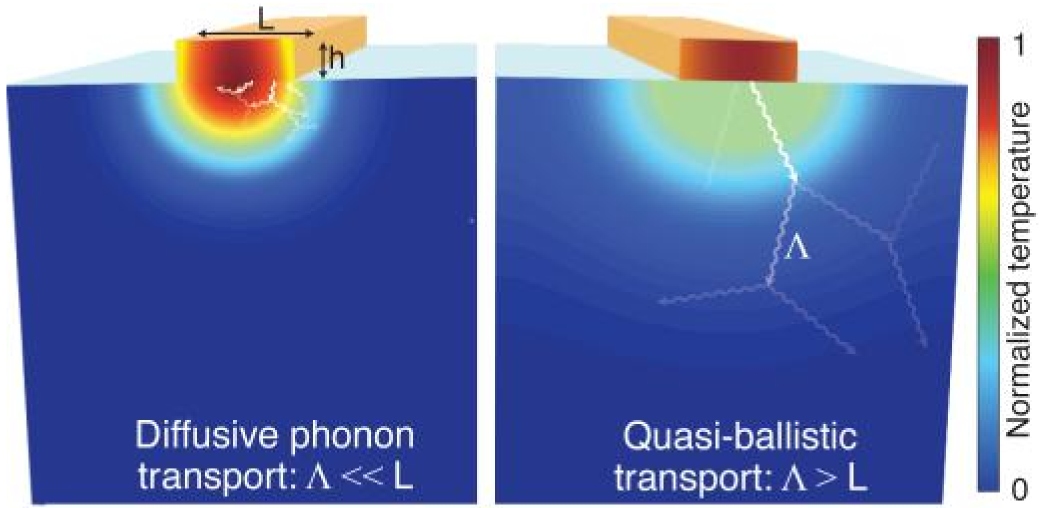
Understanding nanoscale heat flow in 1D

- Heat is carried by phonons
- In the macroscopic world, Fourier Law applies

$$q = -k\nabla T$$

- What happens when a nanostructure is smaller than the phonon mean free path?
- Existing theories of nanoscale heat dissipation disagree
- Fourier law over-estimates the heat flow - need to compare interface dimension to phonon mean free path





Nature Materials ***9****, 26 (2010)*

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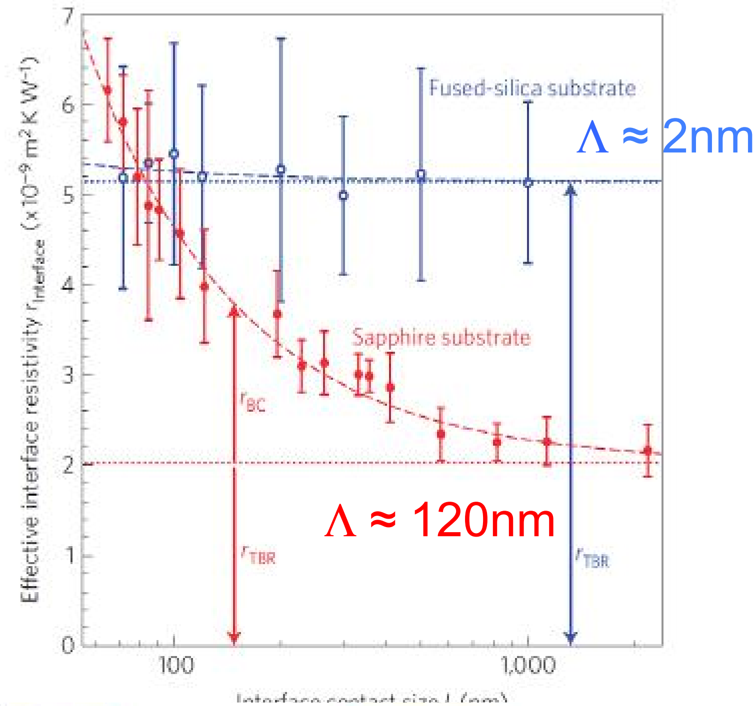
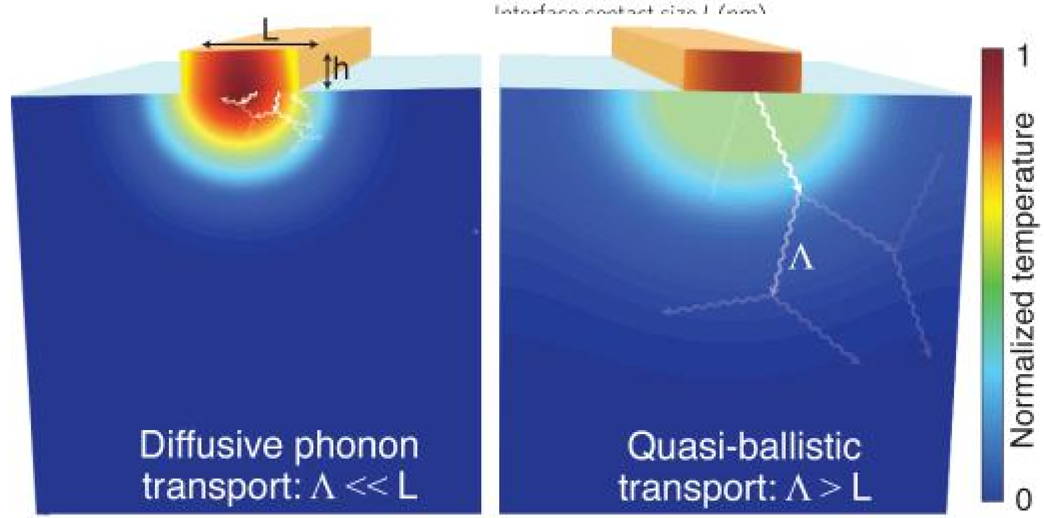


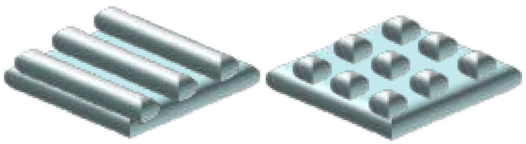


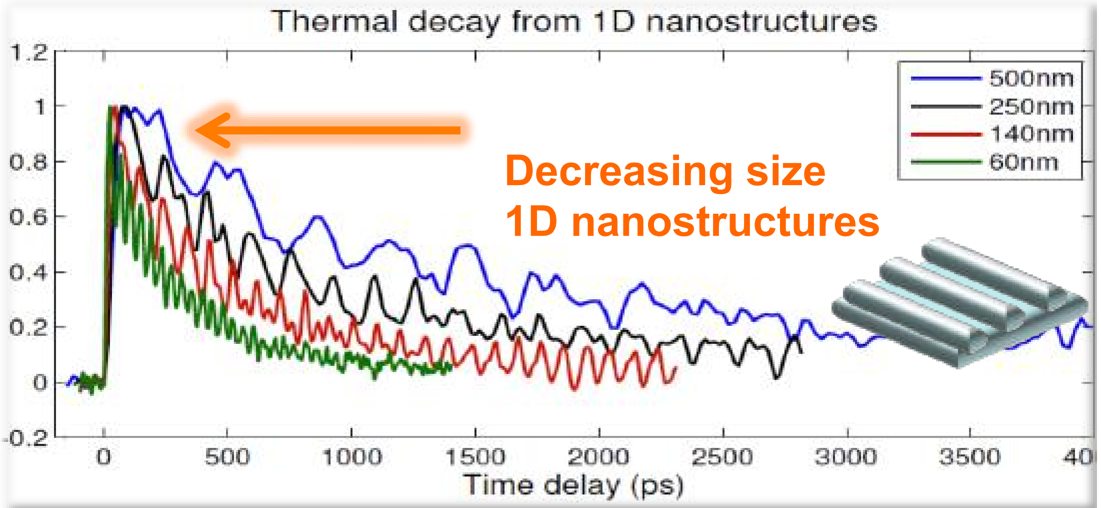
Nature Materials **9**, 26 (2010)

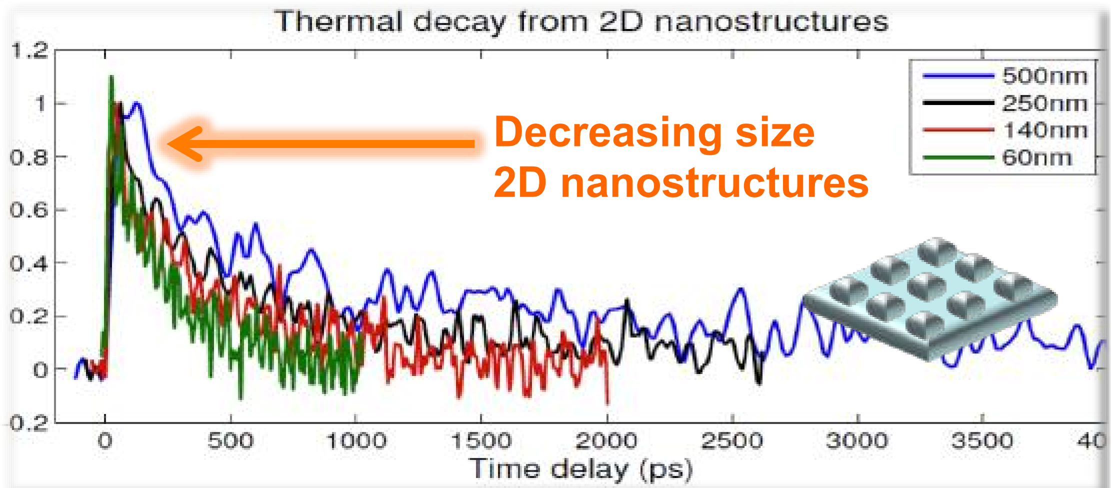
Nanoscale energy flow in 2D even more ballistic

- Thermal decay in 2D slower than 1D on short time scales - stronger ballistic effects!
- At large time scales, thermal decay similar in 1 and 2D - dominated by substrate
- Thermal modeling in progress - no current theory available

Ni on Sapphire







Nature Materials ***9****, 26 (2010)*
Nano Letters ***11****, 4126 (2011)*
PRB ***85****, 195431 (2012)*
Nano Letters ***13****, 2924 (2013)*
Submitted (2013)

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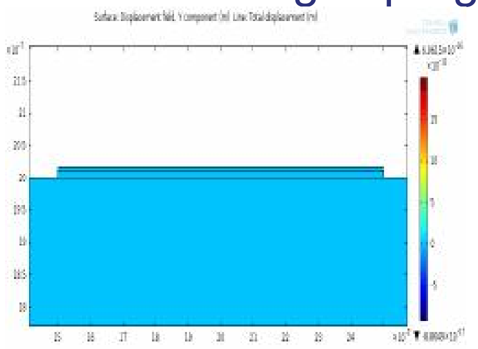
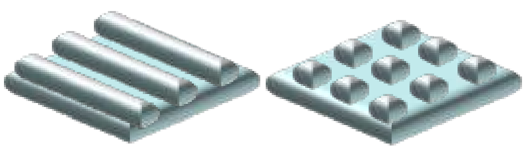
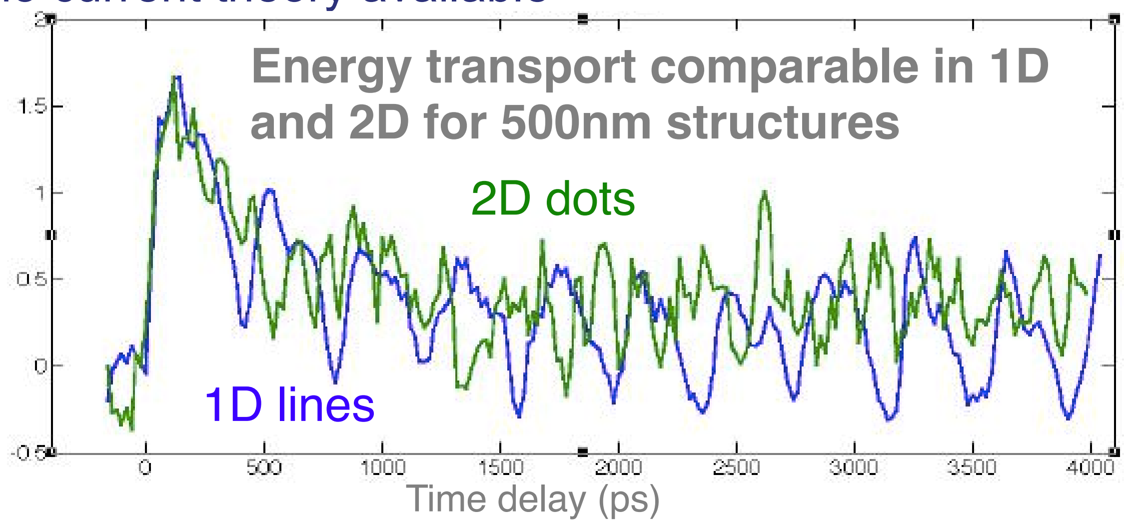
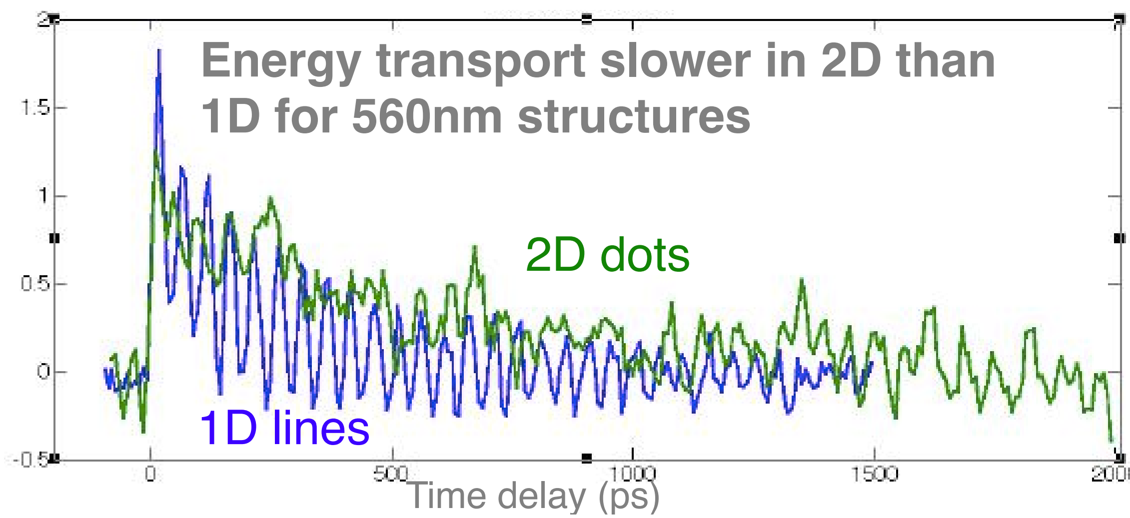


Ni on Si







Nature Materials ***9****, 26 (2010)*
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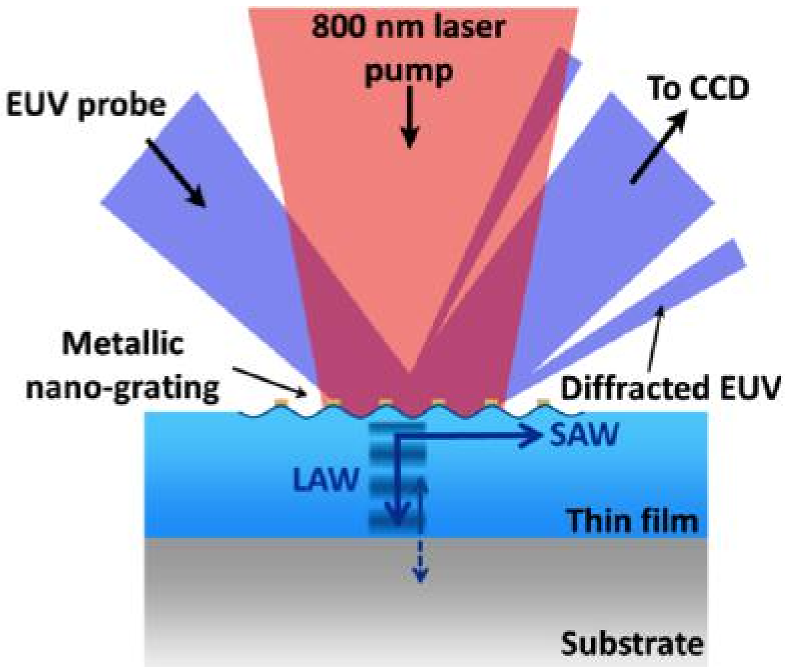
Submitted (2013)

Nanoscale energy flow: acoustics

- Characterizing nanoscale mechanical properties very challenging < 100nm
- EUV HHG is proven to work for < 10nm films!
- Sensitive to pm displacements!
- Demonstrated sensitivity to sub-monolayer!



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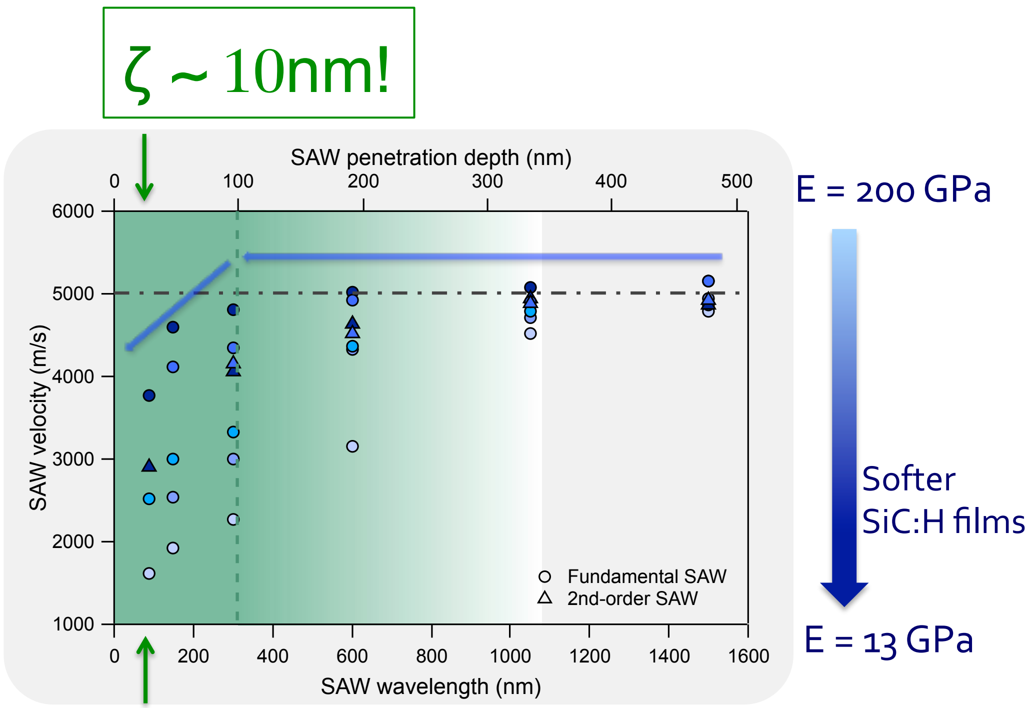
Nano Letters ***11****, 4126 (2011)*
PRB ***85****, 195431 (2012)*
Nano Letters ***13****, 2924 (2013)*
Submitted (2013)



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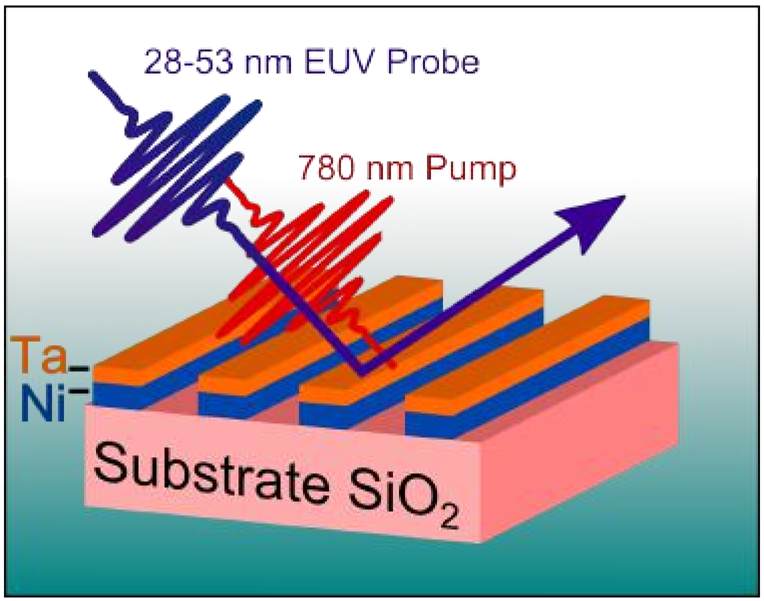
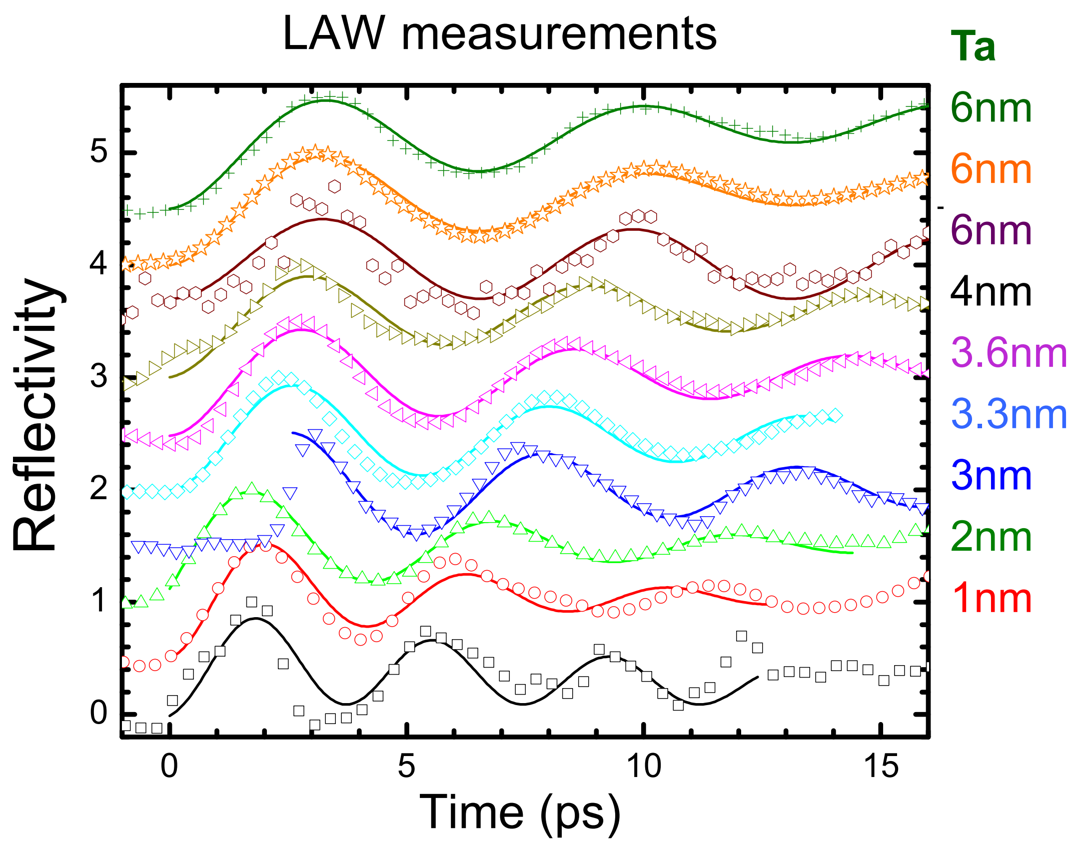
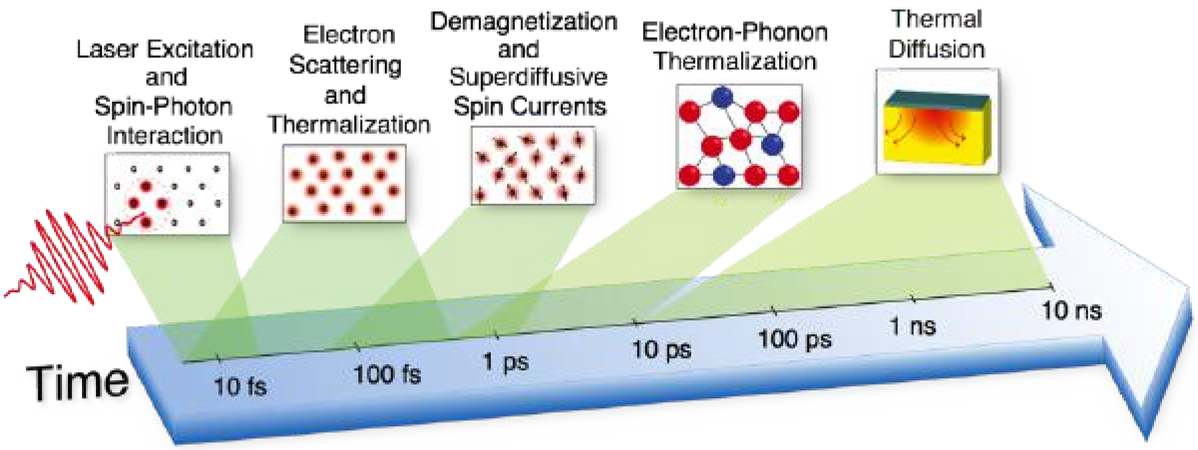
- Characterizing nanoscale mechanical properties very challenging < 100nm
- EUV HHG is proven to work for < 10nm films!
- Sensitive to pm displacements!
- Demonstrated sensitivity to sub-monolayer!
- How do bulk properties develop on layer-by-layer basis?



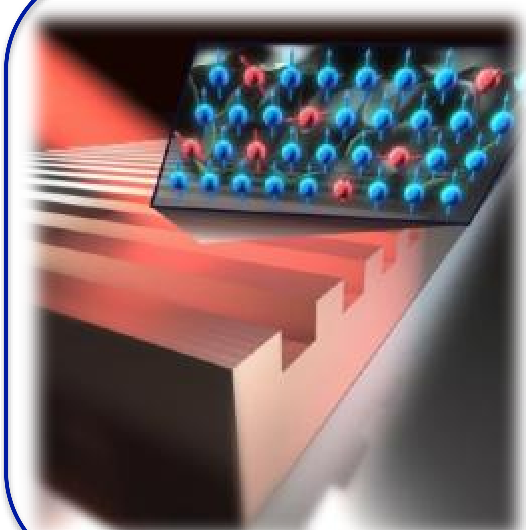


Nature Materials ***9****, 26 (2010)*
Nano Letters ***11****, 4126 (2011)*
PRB ***85****, 195431 (2012)*
Nano Letters ***13****, 2924 (2013)*
Submitted (2013)

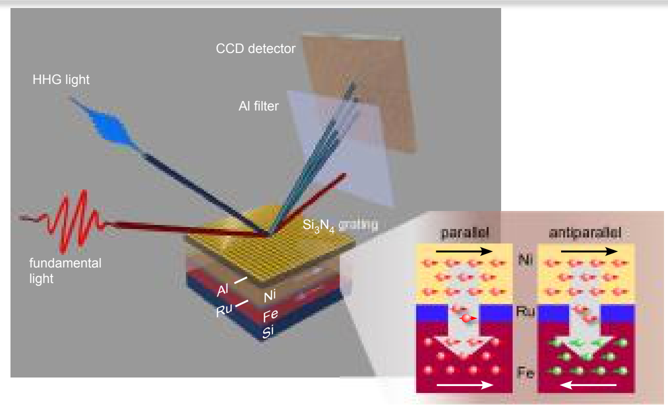
Surprising ultrafast spin dynamics



- No complete microscopic theory of magnetism exists on fs time scales
- High harmonics enable ultrafast, element-specific, spin dynamics to be probed at multiple sites simultaneously



Even in a strongly exchange-coupled Fe-Ni ferromagnetic alloy, the dynamics of the individual spin sub-lattices can be different on timescales faster than that characteristic of the exchange interaction energy (10 – 80 fs)



Large, superdiffusive, spin currents can be launched by a femtosecond laser through magnetic multilayers, to enhance or reduce the magnetization of buried layers, depending on their relative orientation

PUBLICATIONS
*PRX **2**, 011005 (2012); PRL **110**, 197201 (2013)*
*PNAS, **109**, 4792 (2012)*
*Nature Commun. **3,** 1037 (2012)*

NEWS ARTICLES ABOUT WORK
*Physics **5**, 11 (2012)*
*Physics Today **65** (5), 18 (2012)*
*Physik Journal **11**, Nr. 6, page 26 (2012)*

Combine tabletop coherent X-rays with coherent imaging